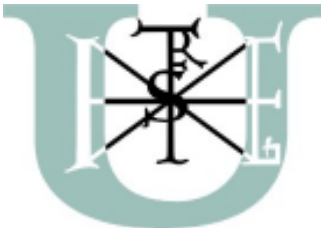

Szent István University

Measurement and simulation of nitrogen exchange between a grassland ecosystem and the atmosphere in landscape scale

The main points of the thesis

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THE BACKGROUND AND AIMS OF THE WORK

Among the elements the nitrogen has one of the most complex biogeochemical cycles; for this reason many transformation processes and factors influencing the N cycle are not completely explored yet. There are some synthesis studies of N-cycles on global scale, nevertheless the uncertainty in estimation of N balance and emissions of biomes remains high because the number of available, appropriate (both lab and field) measurements are relatively sparse. The nitrogen cycle is largely influenced by (not always known) effects of anthropogenic activity, that have impact on concentration, distribution and flux of different reduced and oxidised N-species through affecting the atmospheric chemical processes, as well as metabolic processes of animals, plants and a large variety of microorganisms.

Recently, in evaluation of environmental conditions, balance studies were also published for Hungary beside the simple concentration survey. The trace gas exchange within surface − biosphere − atmosphere system strongly depends on meteorological conditions, concentrations as well as on characteristics of ecosystem and on soil physical, chemical and biological properties. For this reason the investigation of exchange processes above different ecosystems are important and necessary.

The nitrogen budget of the non-intensively managed ecosystems is dominantly determined by the atmosphere − surface exchange processes. For non-fertilized grasslands, the atmospheric deposition is the main source of the nitrogen, but we have to consider the N-fixation by legumes. On the other hand a significant amount of nitrogen compounds is emitted by the biosphere. For various compounds (e.g. ammonia) the exchange is bidirectional, and the sum of deposition and emission rates is the so-called net flux.

The N emission occurs partly from soil and through stomata of plants. In the soil the nitrogen has different forms with large-scale of oxidation number from ammonium (-3) to nitrate (+5). The nitrification and denitrification processes fundamentally affect the soil nitrogen balance, because these processes are producing intermediate gases as nitric oxide (NO), nitrous oxide (N_2O) and elemental nitrogen (N_2), which can emitted into the atmosphere.

The biosphere may emit nitrogen compounds also by the vegetation, dominantly as ammonia gas (NH_3). (Ammonia emission from soils can be observed only for alkaline soils.) The role of ammonia gas in the troposphere, in nitrogen load and in nutrients supply of the ecological systems is well known.

Some of nitrogen compounds have only negative flux (deposition) from atmosphere to the ecosystems, soil and vegetation do not emit them e.g.: nitrogen dioxide, nitric acid (vapour), and ammonium and nitrate particles in fine and coarse mode. Atmospheric gases and particles are deposited by two

ways to the surfaces, partly by precipitation (wet deposition) during the cloud formation and by above cloud scavenging, partly by turbulent flux onto the plant and soil surface (dry deposition). In general, the rates of the dry and wet deposition rates are in the same magnitude.

Above mentioned processes and components determine the surface-atmosphere N-budget of grassland ecosystems. Human activities (both directly and indirectly) may modify the natural N-cycle. Atmospheric lifetime of various N-compounds differs in a large scale − from hour to hundred years − and their environmental impact ranges from local direct damage to climate change.

It is essential to monitor these changes and to determine the rate of pollutant emissions and the harmful effects in current and future context. Although our knowledge is quite comprehensive concerning to these processes there are some uncertainties still remained. Several European research programs (e.g. GRAMINAE, GreenGrass, NOFRETETE and NitroEurope) have been dealing with the nitrogen turnover of various terrestrial ecosystems.

Aims of the study

So far, many nitrogen flux measurements were carried out by different scientific communities from different points of view (e.g., air pollution, greenhouse effect, water pollution, nutrient load etc.), while the number of total N-balance estimations in landscape scale involving all major sources and sinks in total N-flux is sparse.

For detailed investigation the biosphere − atmosphere exchange of different N compounds, including plot measurements over different types of ecosystems and for modelling of N fluxes from plot to continental scale, an EU Framework 6th Integrated Project (NitroEurope) was started in 2006, coordinating the N-researchers across Europe. One of the grassland stations with intensive measurement program of the NitroEurope network was established in central Hungary (Bugacpuszta).

The objective of this work was to investigate the nitrogen exchange between atmosphere and grassland ecosystem and its dependence on climatic conditions, and focusing on links with reactive trace gas emissions and N-depositions, and on the possible feedbacks to soil/vegetation dynamics. In the present Ph.D. work my task was to determine the N-balance between the atmosphere and a semi-natural grassland in a semi-arid continental climate representative for the Hungarian Great Plain, in central Hungary, based on (i) measurements of deposition and emission processes and (ii) using DNDC (DeNitrification DeComposition) model to estimate soil fluxes of N- gases. This work summarizes the results of the five year measurements record and modelling of the N balance, taking into account all the significant N forms.

MATERIALS AND METHODS

Sites of investigations

The selected semi-natural semi-arid sandy grassland site (Bugacpuszta) is considered to be one of the most characteristic landscape types in Hungary. The station is located in the Kiskunság National Park in the Hungarian Great Plain, between the rivers Danube and Tisza. Co-ordinates are 46.69°N, 19.60°E, elevation 113 m above sea level. The climate of the region is semi-arid temperate continental; average yearly precipitation (1989–2006) is 550 mm. Mean annual temperature in this region is 10.7 °C. The soil is a Chernozem-type sandy soil. The vegetation is semi-arid sandy grassland (*Cynodonti Festucetum pseudovinae*) dominated by *Festuca pseudovina*, *Carex stenophylla*, *Salvia pratensis* and *Cynodon dactylon*. The climatic conditions, flora and fauna of the region differ from those in most other European lands. Local plant and animal species have evolved (with extraordinary strategies to survive the sweltering heat and tormenting drought), creating endemic species or subspecies (e.g. Hungarian Grey Cattle). For this reason, the area is a nature reserve and management is not allowed. The plant association is sensitive to disturbances, and the only grazing is by a herd of the ancient Grey Cattle breed (at an average grazing pressure of 0.5–0.8 stock ha^{-1} during the growing season), which has gone on for centuries in dynamic equilibrium with the grass ecosystem.

Description of Denitrification-Decomposition (DNDC) model

A widely used process-based biogeochemical model, DNDC was used to calculate soil fluxes of some N forms (NH_3 and N_2) that are difficult to estimate by field or laboratory measurements. The DNDC model estimates soil fluxes of all important N compounds including NO and N_2O. The model treats nitrogen inputs from atmospheric deposition, fertilizer use and nitrogen fixation and represents soil (in)organic turnover to enable calculation of leaching of nitrogen as well as gas emissions. The model requires many input parameters, including ecological drivers (soil properties, meteorological dataset and farming management), to simulate trace gas fluxes due to the soil climate, and soil processes such as denitrification, nitrification, mineralization, etc. For parameterization and validation of the model, the site-specific soil information (texture, pH, clay fraction, organic C content, bulk density, etc.) was provided by the Research Institute for Soil Science and Agricultural Chemistry (RISSAC) of the Hungarian Academy of Science (HAS) and partly gained by field observations (micrometeorological and botanical dataset) by Szent István University (SzIU). The model consists of two components. The first

component, consisting of the soil climate, crop growth and decomposition sub-models, predicts soil temperature, moisture, pH, redox potential (Eh) and substrate concentration profiles driven by ecological drivers (e.g., climate, soil, vegetation and anthropogenic activity). The second component, consisting of the nitrification, denitrification and fermentation sub-models, predicts NO, N_2O, N_2, CH_4 and NH_3 fluxes based on the modelled soil environmental factors. Classical laws of physics, chemistry and biology, as well as empirical equations generated from laboratory studies, have been employed in the model to parameterize each specific geochemical or biochemical reaction. The entire model forms a bridge between C and N biogeochemical cycles and the basic ecological drivers. The entire model is driven by four major ecological drivers, namely climate, soil physical properties, vegetation, and anthropogenic activities. Sensitivity analyses were also taken to identify which input parameters in the model are mainly responsible for the majority of soil emission change.

Determination of the dry nitrogen deposition based on inferential method

Concentrations of NH_3 gas, HNO_3 vapour and NH_4^+ and NO_3^- particles were measured at 5 m height by daily 24-hour samplings by the three-stage filter pack method (NILU – Norwegian Institute for Air Research – type, applied in the European Monitoring and Evaluation Programme, EMEP network) followed by ion chromatography (nitrate, nitric acid) and spectrophotometry (indophenol-blue method for ammonium and ammonia). The minimum detection limit (MDL) is 0.1 μg N m^{-3} for all components. The precision (bulk relative error) of sampling and measurements was around 10%.

Dry deposition fluxes of NO_2 and NH_3 gases, HNO_3 vapour and NH_4^+ and NO_3^- particles were determined by the inferential method. Dry deposition velocities for different N-compounds onto grass surface, used in the inferential method, were compiled from the literature. Ammonia net deposition (excluding soil emission) was calculated by deposition velocity figures measured above a Hungarian grassland site with similar climate and vegetation/soil characteristics. The slow deposition velocity of NO_2 above grass differs between different sites, depends on climate, has annual and diurnal variations and varies between 0.4 and 2.8 mm s^{-1}. We adopted and used a yearly average deposition velocity (1.35 mm s^{-1}), calculated from literature data for calculation of the dry flux of NO_2. The dry deposition velocity for HNO_3 has been determined previously for a semi-natural grass surface similar to Bugacpuszta. Though there are some differences between species composition, *Festuca pseudovina* dominates at both sites. There are a limited number of estimates for deposition velocities of ammonium and nitrate particles in the

literature. Because deposition velocity depends on the size of particles, mean particle diameter is a good approximation for estimation of v_d. Only a few data are available in Hungary for the nitrate and ammonium particle size distribution. The mean mass diameter of the nitrate particles can be found in the coarse particle fraction (d>2.5 μm), while the ammonium particles are mostly in the fine particle fraction (d<2.5 μm). As the rate of sedimentation (deposition by gravitation) is proportional to the size, the nitrate has a larger deposition velocity than ammonium.

Determination of the wet nitrogen deposition

The wet deposition flux of nitrate and ammonium ions was determined from the concentrations (c) measured in wet-only, daily precipitation samples. Concentrations of ammonium and nitrate ions were determined by ion chromatography and spectrophotometry (indophenol-blue method), respectively. Minimum detection limit (MDL) was 0.05 mg N l^{-1} for both ions. Precision (relative error) of these methods was below 5%. The calculated bulk error of precipitation sampling and concentration measurements was around 10%.

Measurement of atmospheric ammonia gas, nitric acid vapour and ammonium, nitrate aerosol concentrations with DELTA denuder method

DELTA (DEnuder for Long Term Atmospheric sampling) denuder – what we used for the first time in our country – a relatively new, low-cost sampling system, which was originally developed for long-term sampling of NH_3 and NH_4^+, but it is also suitable for sampling acid gases (HNO_3, HNO_2, HCl, SO_2) and particulate aerosol (NO_3^-, NO_2^-, Cl^-, SO_4^{2-} content). Denuder sampling is based on a glass denuder train. The laminar air flow goes through the first denuder tube, where the inner wall of glass is coated with citric acid. The NH_3 is adsorbed on the acidic wall of tube, while aerosol particles pass through the denuder tube due to their inertial mass, and are adsorbed in the surface of a specific back-up aerosol filter. Ammonia and ammonium aerosol separation is achieved by the role that the gas diffusion to the wall is more effective than that of particles. The acidic gases such as HNO_3 were collected by a second alkaline (K_2CO_3) coated denuder in line. The sampling flow rate was between 0.3 to 0.4 L min^{-1}, measured by a high-sensitivity gas meter. The advantage of this method (in contrast with the NILU three-stage filter sampling method) is that the NH_4NO_3 dissociation caused by the pressure drop on the aerosol pre-filter is negligible eliminating both ammonium loss from pre-filter and artefact ammonia on the following, second filter used for sampling of NH_3.

Soil nitrous oxide flux measurements by static chambers

The static chamber method was used to determine the soil N_2O emission. The N_2O soil flux was determined based on weekly sampling by eight static chambers (height 5 cm, area 2,500 cm^2). Collars for the chambers were placed approximately 1 m apart along a 10 m transect. These rims were left *in situ* for the duration of the experiment in order to avoid the high emission peaks frequently observed due to installation disturbance effects. The rims were pushed 4 cm into the soil and were covered by the chamber body only during the 30-minute periods when samples were taken, allowing normal light and precipitation exposure at all other times. Rims were installed to avoid the enclosure of tall plants; short grass within the rim area was not clipped before sampling. Sampling duration was optimized to 30 minutes to eliminate the effect of non-linearity caused by saturation effect. Several pilot measurements justified the linearity of concentration changes in the static chambers above the sandy soil in the first 30 minutes of the enclosure period. Therefore samples were taken at t=0 and 30 minutes after closure of the chambers by syringe into evacuated vials. Linearity tests were repeated once per month. Concentration changes were determined with a HP 5890 II gas chromatograph (Waldbronn, Germany) equipped with a HP-PlotQ column, and electron capture detector (GC-ECD). Calibration and reference gases were provided by Scott and Messer companies (Boulder, CO, USA). Precision and detection limit of concentration measurements were estimated as follows. 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%. CV was determined using at least 10 samples at the start of sampling (t=0). Because samples were taken immediately after closure, concentrations are close to the relatively constant atmospheric background values (320 ppb). The typical CV, during frequent checks of precision, ranges between 4 and 6%. The detection limit was determined taking into account a minimum 10% change in concentration during sampling from the initial background values. According to this criterion, the calculated detection limit of fluxes is 1.3 $\mu g\ N\ m^{-2}\ h^{-1}$.

Soil nitric oxide flux measurements by dynamic chambers

Soil emissions of NO were determined by the dynamic chamber method. Two parallel dynamic chambers (height 5 cm, area 200 cm^2) were used continuously, being moved over 12 different permanent collars in the soil. After a week, chambers were moved over the next two rims, allowing the previous measuring plots to recover. Grass inside the closed chamber was affected by the lack of light and rain for 1 week. Overall, there was no

systematic increase or decrease in NO flux during the continuous enclosure for a week at the same plots. Therefore, it was assumed that this long closure did not greatly influence the soil nitrification processes. After opening the chambers, there was a 5-week period for soil and vegetation to recover. In the calculation of soil flux, the effect of fast chemical reaction between ozone and nitric oxide was taken into account. HORIBA gas monitors (APNA 350 using molybdenum converter and APOA 350) were used to detect the concentration of NO and O_3 leaving the chambers, respectively. The detection limit of the measurements was 0.4 ppb. The precision of the NO/NO_x monitor was tested by calibration gases with mixing ratios of 1 and 10 ppb. The measured coefficients of variation were 5.3 and 1.5% for NO and NO_x, respectively. Because the average mixing ratio during the flux measurements lays between these two figures, our estimated coefficient of variation ranged between 1 and 5%. The overall precision of measurement was around 10%, taking into account the error interval for NO calibration gas provided by the manufacturer. The ozone monitor was calibrated in the central reference laboratory. Calculated precision at the mean atmospheric level (320 ppb) was 2%. Five-minute concentration averages were recorded. Flow rate of the air through the dynamic chamber was 1.5 liters $minute^{-1}$. Due to the short residence time and the steady-state conditions, the rapid chemical gas phase reaction of nitric oxide and ozone, $NO + O_3 \rightarrow NO_2 + O_2$, was taken into account. The photolysis rate of NO_2 inside the dark chambers was estimated to be zero. According to our measurements wall effect was negligible because of the relatively short residence time (40 s) of the gas mixture in the chamber. The inlet for concentration measurement of NO_2 was fixed at 3.5 m above the surface. The ambient air was sucked to a HORIBA (APNA 350) gas monitor through a Teflon tube.

RESULTS

Soil emissions influenced by soil temperature

The NO and N_2O soil emissions as consequence of nitrification and denitrification are, like all biological processes, influenced by temperature and correlate with the soil temperature as many studies discussed before but sometimes the correlation remains poor or understandable. With increasing soil temperature the NO flux increases faster than N_2O up to 20°C; until the role of other factors (e.g. water stress, nutrient supply and these complex processes linked to heat stress) will determine the magnitude of metabolism. The relatively high soil N_2O 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.

It should be noted that the soil temperature cannot explain all of the variations of the soil fluxes. 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.

Soil emissions influenced by soil wetness

The optimum soil wetness for NO and N_2O is ranged around 20-30% and 40-50%, respectively for this sandy soil. The optimum water field pore space (WFPS) content is reached only in 2010 for N_2O. 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. During summer season the microbial productivity is elevated (mineralization, nitrification, immobilization, decomposition etc.) but denitrification (N_2O production) is suppressed when the WFPS is low. In dormant period, despite the higher WFPS the activity of microbial community is decreasing; parallel with soil temperature decrease, resulting in low N_2O production. Changes in seasonality, distribution and frequency of precipitation and the total amount of rainfall may impact greatly on the nitrogen exchange of this ecosystem, resulting in switch between N_2O and NO production (determined by soil processes). Both seasonal and long-term nitrogen exchange of this ecosystem is therefore linked to the soil water content (due to rainfall regime) and soil temperature.

A Dutch research team studied our soils by the 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 for the majority of N_2O production at Bugacpuszta site, thus N_2O can be produced at lower water content – this phenomenon may explain the secondary higher emission on low (20-30%) WFPS. This biochemical pathway also demonstrates that pH may be the major factor determining nitrifier-induced N_2O production and community of micro organism may not be the key driver in different pathways of N_2O formation. By this methodology it was also observed that water is effectively the main oxygen source (instead O_2 as it was assumed earlier) in N_2O formation and possibly for formation of other nitrogen oxides in some European soil samples including Bugacpuszta. The O isotopic measurement of N_2O showed that in Bugacpuszta the soil NH_4^+ can be the N source (and 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 microorganism.

Fluxes of nitrogen components and the N budget

According to the results, the annual precipitation amount in 2007 and 2009 was significantly lower and in 2010 it was two times higher than long term mean. The deficit in the yearly precipitation mainly occurred during July in the vegetation period of 2006-2009 resulting in less biomass production, because the drought and the main growing period coincide. At the same time the lack of precipitation affects the N-cycle through the missing wet deposited N, by the reduced plant uptake, and other soil processes, by water stress etc. In 2007 the annual mean temperature has increased by 1°C associated with mild winter (soil frost was not often occurred) 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 extra precipitated (one of the most wetted in the last 100 years) and differs from the long-term statistical averages. For this reason all the circumstances and conditions are different from the previous years.

Noticeable that the precipitation amount nearly doubled in 2010, nevertheless the wet deposition does not follow this pattern. Ratio of dry to wet deposition varied in the range of 1.5-2.3 between 2006 and 2010.

Based on 5 years measurement in Bugac the average NO_3^- and NH_4^+ content of the precipitation were 2.84 and 0.78 mg l^{-1}, respectively. With except of 2006 the ammonium and nitrate deposition contributes 55% and 45%, respectively, to the total amount of N in the rainfall.

Within the NitroEurope cooperation program the organic nitrogen content of the precipitation samples from October 2008 to February 2009 were analyzed. Based on the results the organic N fraction is about 16% of the total N content of the rainfall, but the short sampling period is not representative, allowing only a rough estimation.

The annual dry deposition of nitrogen compounds of the measurement period was studied and found that the reduced and oxidized forms contribute 55% and 45%, respectively. These results agree with other European measurement sites, although they found slightly larger differences.

The amount of the dry deposition is basically determined by ammonia and nitric acid. It is also noticeable that the contribution of the deposited N gases (NH_3, HNO_3, NO_2) is 83-89%, while the aerosol particles (NH_4^+, NO_3^-) takes only 11-17% in the total amount of dry nitrogen downward flux.

In Hungary we used the DELTA denuder method first time; therefore it was necessary to perform a comparative study with the well known and

accepted EMEP technique. It is an advantage that the DELTA system is operating with low cost due to the monthly sampling, but a disadvantage that we are not able to monitor the daily changes and the extreme values of concentrations. A comparison of monthly data of the DELTA and EMEP technique shows relatively good correlation, except of few months, when the opposite trend are observed, weakening the correlation. Another reason for the difference is that the the two measuring sites lie a few kilometres apart. Yearly course of the NO_3^- flux shows a maximum deposition in winter months, while in summer months the downward flux is significantly reduced. Comparison of the two sampling methods shows a good agreement (r=0.69), even though sometimes the trend turns into the opposite direction. For ammonium results of EMEP versus DELTA denuder methods also show a relatively good correlation (r=0.56). Naturally deposition differences between winter and summer seasons can be observed also for ammonium. In the function of temperature and relative humidity dissociation equilibrium exists among particle NH_4NO_3 and gaseous ammonia and nitric acid preferring gaseous forms in summer half-year.

Ammonia deposition also shows seasonality. In contrast to ammonium, the ammonia dry deposition is largest in summer and smallest in winter. The NH_3 flux has a daily course, which is associated with the compensation point-concentration of the plants. Deposition is higher at night than daytime, when the stomata, are open, but in our case, we could not observed this phenomenon as daily and monthly concentrations were measured by the sampling techniques mentioned above. The two measurement methods showed the weakest correlation (r=0.36) for nitric acid vapour. Nitric acid vapour concentration is generally higher in summer due to the photochemical activity ($HO\bullet + NO_2 + M \rightarrow HNO_3 + M$), and for the reason mentioned in last paragraph but in our case it can not be observed clearly in the annual course.

The multi-year average of the soil N_2O flux measurement is 0.67 kg N $ha^{-1}yr^{-1}$ although there can be large differences between years. This value is close to the mean emission (0.93 kg N $ha^{-1}yr^{-1}$) determined during a survey by nine European grassland measurement sites. The observed lower N_2O emission levels can be exlpained by the differences in temperature, rainfall, by the low N-input, slightly alkaline sandy soil and by differences in water-management features of the Kiskunság area. There is a significant difference between the extremely dry 2007 (annual rainfall: 446 mm) and extremely wet 2010 (967 mm) years.

The N-fixation by plants is not negligible because the share of legumes is 8–17%, according to different surveys, but we were not able to determine the N-fixation by direct measurement. Nevertheless the main N-input of our grassland is the dry and wet depositions (altogether 11–15 kg N $ha^{-1}yr^{-1}$) among sources.

The magnitude of the N deposition is approximately higher by one order than N-emission in this area. If less atmospheric N is deposited to the surface, less mineral N can be leached into the upper layer of soil, thus less trace gasses are produced via nitrification/denitrification processes. As we expected (based on a preliminary study) the NO emission was around 1 kg N ha^{-1} in each year in Bugac station. The NO emission of the landscape of the Kiskunság (Bugacpuszta) area is much lower than for arable (irrigated and fertilized) areas, and slightly higher than for forest. The soil flux of NO is 4-5 times higher in the drier years comparing to the N_2O flux due to the dry soil condition. The soil moisture content of the well-ventilated dry soil is near the optimum for NO formation.

Usually the soil N_2O emission of the fine textured soils is usually higher than for coarse textured soils, while the opposite is true for NO emissions. For this reason the NO emission is low from the loamy soils where emissions of N_2 can be significant. From physical and chemical point of view, these results are meaningful, because the sandy soils have higher permeability than loamy soils. In addition, the advection transport in sandy soil is much more effective and enhances the exchange potential of gases between the soil and the atmosphere compared to molecular diffusion. Therefore, NO from sandy soils is more likely released before it would be involved in chemical or microbial metabolism reaction. The effectiveness of physical way of outgasing from soil is more important for NO than N_2O because the chemical reactivity of NO is higher.

Our observations above slightly alkaline soil match those studies where the ratio of N_2O formation by denitrification is higher in acidic soils than in neutral or alkaline soils. The N_2O emission (<1 kg N $ha^{-1}yr^{-1}$) is considered to be low compared to other results of different measurement sites.

Simulation results

Sensitivity tests were carried out by varying different input factors within the commonly observed range, while keeping all other input conditions to be constant. The main outcomes of the model testing are as follows. Soil pH, SOC (soil organic carbon), precipitation and temperature are the major factors controlling soil processes and gas fluxes. For some components (e.g. for NH_3), soil emission is strongly influenced by soil pH or grazing regime (stocking rate and number of days grazed). There is not direct relationship between emission rate and the temperature because a lot of soil processes are influenced by soil temperature. Depending on the gas component, a 0-25% change of precipitation and ±2°C change of input air temperature give a non-linear answer (variations were between 5 and 44%) for N_2O, N_2, NH_3 emissions and for the emission of the sum of N_2O, N_2 and NH_3 gases. Increasing rainfall,

SOC or temperature results in a higher rate of trace gas emissions and vice versa. The clay fraction plays an important role in pooling the NH_4^+ and has an impact on the nitrification process. N-fixation, grazed biomasses and productivity predicted by the model were not compared due to the lack of measurement or the incomplete database.

Comparing the model results with the measured data

For validation of the DNDC model we compared the simulated NO and N_2O soil flux data with measured fluxes. Monthly fluxes measured by static/dynamic chambers and simulations show a good agreement ($r=0.83$ for NO and 0.74 for N_2O, at the probability level of $p<0.01$). The DNDC model systematically underestimated the higher NO emission peaks. Though the simulation was not able to capture the measured high emission peaks, trends of the monthly emissions give a good agreement. Model simulations of daily soil N_2O fluxes follow well the emission immediately after the rain events. In contrast, by conducting weekly measurements we probably missed some N_2O emission peaks.

In summary, model-simulated soil emissions of the N_2 and N_2O gases, in the period studied, were lower than in regular years because soil conditions did not favour the upper-layer decomposition or the development of anaerobic conditions. The significant proportion of the annual N_2O emission (up to 40-70% of the tundra area) is produced during winter at low soil temperature. The thawing events (when the upper soil layer is not frozen) can cause large emission peaks.

There are several reasons: i) died parts remained in the soil (seed, rhizome, ground-dwelling animals, etc.), which provides additional nutrients (organic C and N) for microorganisms; ii) the lower temperatures is at the optimum for a large group of bacteria; iii) the bacterial diversity (species number and distribution) is an important factor for the share of gases formed.

Seasonal changes of N_2O and NO soil flux can be observed both for measured and modelled values. The explanation is that during summer months the microbial activity of both nitrification and denitrification is increasing with soil temperature.

On the basis of multi-year simulation DNDC model underestimates the emissions of N_2O and NO but the standard deviation of the measured data are much larger than the simulated values. The modelled annual emission level of N_2O is 0.47 kg N $ha^{-1}yr^{-1}$ (which gives a relatively good agreement with our measurements). This value is lower by one order of magnitude compared to the 5.6 kg N $ha^{-1}yr^{-1}$ average value calculated by the IPCC method determined for many European areas. In the IPCC method the cultivated arable lands is also included, where the N_2O emission is generally higher caused by the crop N-

fertilization. There are approximate flux estimations by DNDC model for European grasslands in the literature. Average emission of N_2O by DNDC was estimated 1.01 kg N $ha^{-1}yr^{-1}$. This is in a relatively good agreement with our measured and modelled annual average values, if we consider that the well aerated and dry sandy soil is not favourable for anaerobic denitrification producing N_2O.

It can be concluded (based on the model results) that the rates of annual soil N emissions vary in a relatively narrow range, with an average of 2.0±0.44 kg N $ha^{-1}yr^{-1}$ in Bugac. There are significant differences between shares of different N-components in the given year, which was influenced by change in meteorological variables.

Soil emissions of N_2 and NH_3 have not been measured because of practical reasons. Hence, validation of the model for these parameters was not possible due to the lack of measurement. However, taking into consideration the relatively good agreement between modelled and measured fluxes for NO and N_2O, we can make a rough estimation (assuming a similar relationship) for N_2 and NH_3 fluxes, while noting that the uncertainty of these values are higher than for other components.

Based on the simulations the annual average of the N-removal by grazing is 372±87 kg C and 20.8±4.8 kg N per hectare which shows very good agreement with the calculated data. With grazing the nitrogen temporarily leaves the ecosystem and it is practically supplied back with excreta except the amount of nitrogen (~4 kg N $ha^{-1}yr^{-1}$) built in bodies of cattles.

Overall, the results of the simulation usually match the measured monthly mean measurement values, but in some cases the fluxes are over- or underestimated by the model. One should not forget the three-dimensional heterogeneity of the ecosystem (even on meter scale) it was the reason that measurements were taken at several places simultaneously. The microbial activity often responds much quicker to the environmental changes, such as biomass, but the latter remains an important character. The major simulated trace gas emissions and short-term events are often driven by:

- meteorological extreme events (heat and water stress, freezing-thawing),
- drying/rewetting (nitrification-denitrification),
- management (grazing/cutting/fertilization etc.).

New scientific results

1. Based on measurement and model simulations for a semi-arid semi-natural grassland I quantified the dry and wet N depositions and soil N-trace gas fluxes produced by microbiological processes in different years. Without anthropogenic activity the ammonia deposition is the main N-source (35-40%) of the area. The dry deposition of nitric acid vapour (20-25%) and wet deposition of N (30-35%) is also significant. In European relations the N-gas emission (2.0±0.4 kg N $ha^{-1}yr^{-1}$) of the area is not considered to be significant. The amount of nitrogen load is also less than in other European sites due to the lack of fertilization.

2. I demonstrated how deposition of N-components and the relative ratio of the N-gas emissions are influenced by different weather conditions like precipitation, and temperature trends year by year. The annual rainfall (in contrast as we expected earlier) was not significantly affected the share of wet deposition in the total N-input (which ranges between 30-40% in each year).

3. I described the relationships between soil temperature and soil moisture, and the NO and N_2O fluxes for the sandy soil, which slightly differs from other examined soils. By the increase of soil temperature the biological activity (nitrification and denitrification) is also increasing since trace gas emission intensity also increases up to ~20 °C until other factors (substrate availability, or soil moisture content etc.) do not limit gas production. The intensity of gas formation is highest when the soil moisture content is 20-30% and 40-50% for NO and N_2O respectively. Due to the properties of sandy soil, generally the nitrification dominates, which favours the formation of NO. The soil becomes anaerobic after rain events, thus the significant emission peaks of N_2O and N_2 by denitrification can be observed only for short periods. The rate of N_2O/N_2 production depends on WFPS through the amount of precipitation. An alternative metabolic process (namely the nitrifier denitrification) of ammonia oxidizing bacteria controls the formation of N_2O at lower humidity levels in soil, resulting in a secondary peak in the range of lower wetness.

4. Based on the measurements and model calculations I estimated the nitrogen budget of Bugac landscape in different (dry and wet) years. The Bugac (nature reserved area) representing a background levels of fluxes caused by the absence of local air pollution or N-sources (fertilizers etc.). By averaging many years, the gross N deposition is 13±2 kg N $ha^{-1}yr^{-1}$ and the emission is 2.0±0.4 kg N $ha^{-1}yr^{-1}$ at this grassland, thus the net

flux varies between 9-13 kg N $ha^{-1}yr^{-1}$ in each year depending on meteorological conditions. The net N consumption by Hungarian gray cattles is 4-10 kg N $ha^{-1}yr^{-1}$ is comparable to the net N-exchange (between soil and atmosphere); therefore the N budget of this landscape is nearly balanced, considered to be a slight N-sink.

5. I built up a database of the air, soil and other parameters for an ecological model. In Hungary I applied and validated first the DNDC model using the measured data of Bugac.

6. Soil processes of the sandy soil were simulated in different years and the results were compared to the measured soil fluxes. There are good agreement between measured and simulated annual soil N fluxes and C/N ratio of soil and plants. The correlation of measured and modelled N_2O and NO monthly soil flux data is r=0.8 and 0.7, respectively. The measured and simulated average C/N ratio of the soil and vegetation is 14.3; 12.6 and 20.6; 19.0 respectively.

CONCLUSIONS AND RECOMMENDATIONS

Seasonal fluctuation of N_2O and NO emission has been mainly influenced by precipitation. N_2O emissions were not significant (<1 kg N $ha^{-1}yr^{-1}$) in the N budget at this site in the last few years. Reduced N_2O emission (occurring desertification due to perturbed climate conditions) means a potential negative feedback to greenhouse effect. On the other hand the vegetation can turn into being a net CO_2 source in extreme dry years like 2003 and 2007 as a positive feedback for climate change. The ratio and strength of this two phenomena cannot be neglected due to 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 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 N_r loss by fires is equals to or even exceeds the amount of nitrogen from atmospheric deposition.

It seems that Bugac as a nature reserve (low atmospheric input, without other substantial N source such as N-fertilizer, manure or N-fixation, and with

only extensive grazing management and low air pollution levels) represents a close to the background region for reactive N-fluxes at the regional scale.

These results, together with other studies for grasslands, show that European temperate grasslands act as a weak net sink for atmospheric N, and that observed soil emissions are lower by one order of magnitude than atmospheric deposition. Wet and dry (mainly NH_3) depositions dominates in the N-balance during the observed period. Soil trace gas emissions are strongly controlled by soil organic carbon (SOC) and soil mineral N-content, and by soil temperature and moisture.

Weather perturbation can substantially modify both the timing and the magnitude of N-deposition and soil N-gas emission. In summer time parallel with the precipitation deficit less easy available N is deposited to the surface and leached 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, lots of 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 N gas emission rates, though the complex system of the relationships makes difficult to explain all changes of NO and N_2O formation. Though NO flux was higher than N_2O flux, the soil emission was an order of magnitude lower than atmospheric deposition.

Based on the observed phenomena, it can be concluded that climate extremes is a significant factor in the soil organism functioning and dynamics of N-exchange and emissions. Hence, N-content of the soil is continuously changing with the climatic anomalies. Due to the pool of the NH_4^+ and NO_3^- – which depends on the N-consumption and exchange of soil microbial community – affects the plant N-uptake (demand), plant growing etc. The living roots and bacteria are competitors for the same nutrients, so plants also induce effect on soil N transformation. Further research relating to soil biochemistry in natural grasslands is also needed. Compared to the average of earlier years (13.8 kg N ha^{-1}) less N deposited onto the surface (11.2 and 11.7 kg N ha^{-1}) in the driest period (2006 and 2007) and more N deposited (15.0 and 14.5 kg N ha^{-1}) in the wetter years (2008 and 2010) during the studied period. Wet deposition of nitrate and ammonium together with the dry flux of ammonia and nitric acid vapour 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 the precipitation, soil moisture, air and soil temperature as easily measured parameters more or less can explain this variability.

Rate of atmospheric N deposition determines the rate of N losses. Low N deposition rates decreases N loss as N_2O and NO emissions and NO_3^- leaching. The share of N species in the loss depends on soil properties, climate and

ecosystem type. Using the measurements and model simulation the calculated N budget between the atmosphere and the surface at the examined semi-arid, semi-natural, protected grassland between 2006 and 2010 ranged between -8.8 and -12.5 kg N $ha^{-1}yr^{-1}$ as the sum of the estimated deposition and emission terms: -11.2–(-15) and 1.4–2.9 kg N $ha^{-1}yr^{-1}$, respectively.

The sandy soil at our site was extremely dry, with an average water-filled pore space of about 40% during the examined period. In the dry soil, the denitrification process was limited because soil wetness was usually lower than optimum for nitrous oxide production and following emission. Due to the irregular weather during the study period (2007 was less precipitated and 2010 was more precipitated), all N fluxes were different to those usually found.

Wet and dry (mainly NH_3) depositions dominated the N balance during the examined period in the Hungarian semi-arid grassland. Soil trace gas emissions are strongly controlled by soil organic carbon (SOC) and soil mineral N-content, and by soil temperature and moisture. These parameters are treated by the DNDC model. The DNDC model can simulate the seasonal patterns of N_2O, NO, NH_3 and N_2 soil fluxes. The daily flux simulations usually matched the measured data for N_2O and NO, though in some cases the mean fluxes were over or under-estimated. The results of the comparison of the monthly values suggest that the model, together with the observed deposition data, is applicable to the grasslands to estimate the net N balance. Using the DNDC model we are able to give N-gas flux prediction for those lands where measurements are missing and we are able to simulate fluxes of parameters and soil processes where field or laboratory measurements are difficult or expensive. This provides some support for future use of the DNDC model in regional mode for scaling up the soil fluxes for different ecotypes or give climate scenario estimation up to country scale (using GIS database). In Hungary the rough estimation of greenhouse gas emissions by IPCC could be refined using the DNDC.

The accumulated reactive nitrogen (N_r) in the soil can lead to productivity growth, or loss of biodiversity. The residence time of N is potentially lower than untreated grasslands. The application of chemicals and fertilizers in sandy grass may temporarily lead to yield growth, but the original vegetation can be irreversibly destroyed. The selection of herbivore is important, because every species have plant preferences. The optimum number of animals (0.6-0.8 animals ha^{-1}) is also required to preserve the treeless condition and plant species diversity through small disturbance. With overgrazing the nitrogen can be mobilized faster in the soil due to manure and urine and may lead faster biochemical processes. The compaction of the upper soil layer due to animal trampling can cause airless condition (decreasing porosity). This may increase the denitrification activity, which can lead to increased N losses.

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