DYNAMICS OF NITROUS OXIDE EMISSION OF A TYPICAL AGRICULTURAL REGION

Final report on OTKA K109764 project

Principal investigator: Haszpra, László

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Introduction

Nitrous oxide (N_2O) is one of the most important greenhouse gases (GHGs) directly influenced by anthropogenic activity. Under our conditions, its waste majority is emitted by the agricultural sector where the emission from soil is sensitive to the environmental conditions. In Hungary, N_2O emission is known with the highest uncertainty among the major GHGs. Calculation of N_2O emission is based essentially on statistical methods, theoretic emission factors, and the resulted uncertainty is well above 100 % (Hungarian Meteorological Service, 2017). The sporadic emission measurements applied momentary point sampling (chamber methods with GC analyses of a limited number of air samples), and they could not reveal the dynamics of the underlying processes. The technological development in the last few years have made it possible to continuously monitor the N_2O emission with high precision and fine temporal resolution, however standard methodology has not been developed yet. The spatial representativeness of the measurements depends on the location of the monitoring system above the ground. So far, only low elevation (2-10 m above the ground) monitoring systems have been put into operation in the world focusing only on the underlying specific ecosystem (see e.g. Haszpra et al., (2018) for review). Upscaling of their results for landscape or larger scales required by the regional and global greenhouse gas budget models is difficult and uncertain. Therefore, measurements on tall towers providing larger spatial representativeness would be desirable.

Aim of the project

In the framework of the project, we intended to design a monitoring system applying the state-of-the-art eddy covariance technique, which can be mounted on a tall tower and can monitor the vertical flux of nitrous oxide (emission from the surface to the atmosphere) with high precision on long-term. The system was intended for the Hegyhátsál tall tower greenhouse gas monitoring station (Vas County) located in an unpolluted rural environment and surrounded by agricultural fields, where the dynamics of the nitrous oxide emission of an agricultural area can be studied. By the end of the project, we expected a flux data series, which gave us the first information on the real nitrous oxide emission of an agricultural area and from which the basic features of the dynamics of the nitrous oxide emission could be revealed. The intention was to construct a monitoring system that could be used for different research purposes in the future without further significant investment.

Difficulties and changes during the project

At the time of the preparation of the financial plan for the proposal and at the time of the ordering of the N_2O gas analyzer manufactured in the United States the USD/HUF exchange rate was around 220, while by the time of the delivery of the instrument, when the invoice had to be paid, it increased by more than 10 %. This price-explosion upset the financial balance of the project, where the cost of the analyzer was originally planned to add up as much as 63 % of the total net project budget. Due to the resulted significant financial deficit in the first project year, the monitoring system could be completed only by the middle of the second project year, almost a year later than originally planned. Even the original measurement period would have been rather short for scientifically sound results, the loss of almost one year would have made the original goals unachievable. Therefore, we applied for a one-year prolongation of the project, which was granted.

Results

In the framework of the project, we have constructed a unique, dual-purpose monitoring system capable to measure both the atmospheric concentration of nitrous oxide with the precision required by the international monitoring networks (WMO, 2016) and its vertical flux characterizing the nitrous oxide emission of the surface (vegetation+soil) to the atmosphere. In this way, the monitoring system can satisfy the demands of both the "bottom-up" and the "top-down" N_2O atmospheric budget models. The core element of the monitoring system is a Model 913-0014 Enhanced Performance fast-response N_2O/CO/H_2O gas analyzer manufactured by Los Gatos Research Ltd., San Jose, CA, U.S.A. The monitoring system has been installed at

82 m above the ground on a telecommunication tower at Hegyhátsál, which was already the base of several greenhouse gas related monitoring and research projects. The exceptionally high elevation location of the system provides large spatial representativeness. Here, the footprint (also called as source area) of the measurements covers an agricultural area, where the non-biotic (e. g. fossil fuel originated) nitrous oxide emission is negligible. As the measurements characterize a more or less typical mixture of different agricultural fields, the results can be generalized, upscaled for larger regions with less uncertainty than the common measurements focusing only on a single ecosystem. According to our best knowledge, the only other tall tower nitrous oxide flux monitoring project in the world was initiated in Denmark (Ibrom et al., 2015), but it was suspended for technical reasons before the publication of any data (A. Ibrom, personal communication). The nitrous oxide flux monitoring system at Hegyhátsál has been combined with the long-existing eddy covariance system for carbon dioxide (CO_2) flux monitoring system (Haszpra et al., 2001), and so, now, the measuring system is capable to provide synchronized nitrous oxide, carbon monoxide, and carbon dioxide flux data, which offers exceptional research possibilities in the future.

Hegyhátsál is also a cooperative site of the National Oceanic and Atmospheric Administration Earth System Research Laboratory (NOAA ESRL, U.S.A.) Cooperative Global Air Sampling Network (https://www.esrl.noaa.gov/gmd/ccgg/about.html, NOAA station id.: HUN) where a pair of flask air samples is taken for greenhouse gas measurements every week in the early afternoon hours when the atmospheric mixing is the most intensive. The air samples are shipped to NOAA and they are analyzed at its laboratories in Boulder, Colorado, among others for nitrous oxide. The comparison of our in situ measurements with those absolutely independent ones has proved that the precision of our system satisfies the compatibility goal of the World Meteorological Organization (WMO, 2016), as the average deviation between our measurements and the global reference scale is <0,1 ppb (in fact, the average deviation is only 0,08 ppb).

Nitrous oxide surface-atmosphere flux (emission) monitoring by means of eddy covariance technique is globally rare, and tall tower application has not been published yet at all. Therefore, the complete methodology including the quality control and data gap filling procedures have to be developed. The methodology elaborated is partly based on the general methodology of eddy covariance measurements, and partly on the experiences gained during the present project.

According to the traditions in this field of science, data series for complete calendar years are evaluated to follow the biological cycle of the vegetation. As the measurements started in summer, 2015, data series for a full calendar year became available at the end of 2016, but it seemed reasonable to wait with the data evaluation until summer, 2017, to have two complete meteorological cycles. The ‘summer 2015 – summer 2017’ data series allowed us to give a realistic trend estimation on nitrous oxide concentration, not distorted by an incomplete annual cycle. Our measurement shows a 0.91 nmol mol^{-1} $year^{-1}$ increase in the atmospheric concentration, which is almost identical with the global average in the recent years (0.90 nmol mol^{-1} $year^{-1}$ - WMO, 2017). The measured average N_2O concentration was 330.7 nmol mol^{-1} at Hegyhátsál in 2016, which exceeds the global average of 328.9 nmol mol^{-1} (WMO, 2017) only by less than 2 nmol mol^{-1}. The positive difference indicates the continental/European origin of the surplus N_2O, while its low magnitude reflects the long atmospheric lifetime of N_2O that allows the fairly homogeneous mixing of the emission in the global atmosphere.

Due to instrument malfunctioning, power failure, maintenance, calibration and other technical reasons 17.2 % of the potential data were lost. During the quality control process, a few more data were also flagged as bad or suspicious. The total data availability for the two years long measurement period was 82.2 %, which is a fairly high value relative to the common eddy covariance measurements. N_2O flux was calculated on hourly base. Daily flux can only be calculated if all 24 hourly data are available for summing up, and annual flux can be calculated if daily data are available for all days of the year. If any data is missing the data gap has to be filled for the calculations. Short gaps (<3 h, typical for maintenance and calibration) was filled by means of linear interpolation between the available data, while for longer gaps the median flux for the given month and for the given time of the day was applied.

During the 1 July, 2015 - 30 June 2017 period the total net nitrous oxide release from the surface was 441±195 mg N_2O-N m^{-2}. For the calendar year of 2016, it was 221 ± 141 mg N_2O-N m^{-2} (equivalent to 347 mg N_2O m^{-2} $year^{-1}$). The emission shows a weak annual cycle with a minimum in winter, when the microbiological activity in the soil is minimum, and a maximum in early summer, when the spring fertilization and the favorable environmental conditions result in high N_2O production. An interesting special feature of the emission is that a significant part of the total emission is released into the atmosphere during short (a few days) spring-summer periods after heavy precipitation (>20 mm day^{-1}). It indicates that the soil water content plays a governing role in nitrous oxide production, in addition to the temperature, soil nitrogen content etc.

Nitrous oxide uptake by agricultural soil is unlikely, but it is still an open question (Chapuis-Lardy et al., 2007). We measured negative fluxes only winter and within the uncertainty range of the measurements. Due to the low soil temperature during this season biological activity in the soil is limited, therefore the occasional negative fluxes are likely only the results of the measurement uncertainty. Our results support the common assumption that agricultural soils are rich enough in nitrogen not to uptake nitrous oxide from the atmosphere.

On the basis of the measurements, we have started to develop a process-oriented biogeochemical model to describe the surface-atmosphere nitrous oxide flux as a function of the environmental factors. The present version of the model performs well but still more data, longer measurement data series are needed for fine-tuning and validation.

Publication activity

The workplan of the present, essentially infrastructure-oriented project promised a single publication, preferably in a high-class international journal, at the end of the project summarizing the structure of the monitoring system developed and its first results. The description of the monitoring system and the results listed above have been published in Atmospheric Environment, in a Q1 category international scientific journal, at the end of the project (Haszpra et al., 2018). Taking into account the pioneering nature of the work, we expect a high number of citations in the coming years. After the start of the measurements, in the second half of the project, the scientific community was informed about the progress in the project, about the preliminary results on posters presented on international conferences (see the publication list). These posters also presented the first results of our modeling activity, which are not mature enough for a regular publication yet.

Future research possibilities

In this project, most of the time and the resources were taken by designing and installation of the measuring system that allowed only a narrow focused research and only the evaluation of part of the data produced. However, the combined N_2O/CO/CO_2 monitoring system constructed offers a wide range of research possibilities without further major investment. Among others we can develop sophisticated quality control methods to improve the quality of flux measurements of any of the gases, we can improve the emission factors for fossil fuel and biomass burning, we can attribute the fluxes to different vegetation types, we can map the regional emission of these components, and having a longer data series we can get more information on the dynamics of nitrous oxide emission of agricultural regions. All these topics will be included in our future project proposal to take the advantage of the infrastructure built and to improve its cost/benefit ratio.

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