

16. Biosphere-Atmosphere Interactions and Greenhouse Gases

+

17. Atmospheric Chemistry and Physics

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16.1

Quantification of methane emissions from a waste water treatment plant in Switzerland

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Quantification of gaseous emissions from waste water treatment plants (WWTPs) is challenging due to the heterogeneity of the emissions in space and time. The inverse dispersion method (IDM) using concentration and turbulence measurements in combination with a backward Lagrangian stochastic (bLS) dispersion model based on Flesch et al. (2004) is a promising option to quantify such emissions. It is increasingly used to determine gaseous emissions from confined sources (Flesch et al., 2009; VanderZaag et al., 2014), as it offers high flexibility at reasonable costs. For the application on WWTPs the bLS model assumption of spatially homogeneous turbulence, which implies absence of obstacles as buildings and trees that disturb the flow, is often not fulfilled. However, studies showed that with the correct instrument setup and data filtering the bLS can be used for emission estimates. In this study methane emissions from a WWTP in Switzerland were quantified using the IDM with the bLS model. Methane concentrations were analysed with open-path tunable diode laser spectrometers (GasFinder3-OP, Boreal Laser Inc., Edmonton, Canada) placed up- and downwind of the source. Here we present WWTP emission estimates from the 20 day measuring campaign.

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16.2

New steps towards a model-based estimation of N₂O emissions from agricultural soils in Switzerland

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The agriculture sector is responsible for almost 13 per cent of all greenhouse gas emissions in Switzerland (FOEN 2020), with a significant contribution of nitrous oxide (N₂O)-emissions from agricultural soils. The amount of nitrogen converted to N₂O remains highly uncertain due to the very dynamic nature of soil processes that lead to N₂O production. Because of the complexity and the need of detailed databases covering the whole territory of a country, including reliable weather and farming data, so far only few countries have been able to use modelling for their national N₂O-inventories of the agricultural sector.

In the present study, we evaluate the process-oriented model DayCent for estimating soil-based N₂O emissions from agricultural soils in Switzerland as a first step towards implementing a more detailed methodology for national greenhouse gas reporting. To test the model performance, we are comparing the model outputs with soil N₂O fluxes measured in field experiments. A selection of field studies from the literature was performed considering the following criteria: (i) the measurements of N₂O emissions were performed under pedoclimatic conditions and agricultural management representative for Switzerland; (ii) studies with a reliable strategy of N₂O monitoring (e.g., number of measurements over time); (iii) with measurements performed for at least one year; (iv) at least one important auxiliary variable related to N₂O emission was analysed, including soil water-filled pore space (WFPS) and soil mineral N content.

After a preliminary tuning of the model parameters, the results of the simulations showed, in general, a proper capability of the model in predicting the management-induced effects on soil N₂O fluxes. DayCent simulated quite well the tillage-generated mineralization of SOM resulting in production of mineral N and soluble C and triggered, consequently, the N₂O production from soil indigenous N due to denitrification. The soil WFPS, which indicates the soil aeration status and so the potential for denitrification-induced N₂O emissions, was in acceptable agreement with the measured values.

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16.3

Modelling current and future atmospheric selenium deposition using available observational constraints

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Selenium (Se) is a trace element that is essential for humans and animals to have in their diet for proper physiological functioning. Between 0.5 to 1 billion people are estimated to have inadequate dietary Se intakes. The concentration of Se in crops depends on the amount of bioavailable Se in agricultural soils, which in turn depends on the amount of atmospheric Se depositing on these soils. However, until now, very little was known about the atmospheric Se cycle in general, and past quantitative estimates of global atmospheric Se fluxes were highly uncertain. Therefore, we implemented the Se cycle in a three-dimensional aerosol-chemistry-climate model, SOCOL-AER, to simulate the transport and deposition of Se. This model can be used to identify areas where atmospheric inputs to soil Se are low and predict future trends in Se deposition.

The Se cycle in SOCOL-AER is based on the existing sulfur (S) cycle in the model, because these two elements have similar atmospheric sources and chemical properties. Gas phase Se species (DMSe, H₂Se, CSSe, CSe₂, OCSe, and SeO₂) are emitted by natural processes — including volcanic degassing, the marine biosphere, and terrestrial biosphere — and anthropogenic activities — including coal combustion, metal smelting, and biomass burning. Once in the atmosphere, the volatile Se species are quickly oxidized (on timescales of minutes to hours), forming low volatility compounds that partition to the particulate phase. Particulate-bound Se is transported on average for around 4.5 days in the atmosphere and is ultimately removed through wet and dry deposition.

In order to constrain the atmospheric fluxes of Se, we compare the new Se model with a compiled database of more than 2000 aerosol Se measurements using Bayesian inference methods. When optimized emission parameters are used, SOCOL-AER shows reasonable agreement with the aerosol Se measurements included in the optimization ($R^2 = 0.66$), as well as with independent aerosol ($R^2 = 0.59$) and wet deposition measurements ($R^2 = 0.57$). We find that 29 and 36 Gg of Se cycle through the atmosphere every year, which is around double previous estimates. Atmospheric inputs of Se to agricultural soils and the marine environment may therefore be larger than previously expected. Currently, anthropogenic emissions contribute around 35% of total Se emissions. However, anthropogenic emissions of Se have decreased over the last three decades in North America and Europe, due to changes in technology and energy policy.

In the future, anthropogenic S and Se emissions are expected to decrease further because of improved pollution control technology and shifts away from coal power generation. To investigate the impact of these changes on agriculture, we model the changes in S and Se deposition between the 1980s and the 2090s in SOCOL-AER under two socioeconomic scenarios. Sulfur and Se inputs to soil in the Northern Hemisphere are significantly reduced (–55 to –90%) by the end of the 21st century. Climate change will alter precipitation patterns in the future, though this effect on Se deposition is minor compared to projected emission changes. While the decrease in coal combustion is a success for environmental protection and climate change mitigation, it may lead to consequences for industrial agriculture. Due to reduced S and Se supply from the atmosphere, fertilization and biofortification strategies will need to be developed to prevent S deficiencies in agricultural crops and Se deficiencies in humans and animals.

16.4

Investigation of the Nature and Variability of Ice Nucleating Particles to Understand Ice Formation in Arctic Mixed-phase Clouds

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As climate change has an amplified effect in the Arctic regions, it is crucial to better understand the properties of Arctic clouds. Aerosol particles significantly influence cloud properties such as lifetime, optical thickness and cloud phase, by acting as cloud condensation nuclei and ice nucleating particles (INPs). These properties are instrumental to understand the radiation budget and hydrological cycle, which are key for climate projections. The concentration of ambient INPs in clouds, responsible for the initiation of atmospheric ice formation at temperature > -38 °C is highly uncertain and variable. So far, the understanding of the origin and nature of INPs in the Arctic remains unclear owing to the scarcity of data and the low overall aerosol concentrations that challenge INP measurements.

In this study, we performed two field campaigns in autumn 2019 and spring 2020 in Ny-Ålesund, Svalbard (78.92°N, 11.91°E), where we monitored ambient INP concentrations continuously at -30 °C in the immersion freezing mode using an online continuous flow diffusion chamber (HINC, Lacher et al., 2017) at a temporal resolution of 20 minutes. Additionally, we deployed a PM₁₀ filter collector at a sampling interval of 8 hours to perform offline analysis of INP concentrations at temperature > -22 °C via an offline drop-freezing technique (DRINCZ, David et al., 2019). Aerosol properties including size distributions and the contribution of fluorescent particles as a proxy for biological aerosol were also monitored for determining the source of aerosol particles throughout the measurement period. We will report the results combining the above measurements, which allows determining the INP concentrations, their variability, nature, and contributions to primary ice formation in Arctic mixed-phase clouds. In particular, we will present our findings on the role and connection between biological sources and INPs in the Arctic.

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16.5

Top-down Estimation of Swiss non-CO₂ Greenhouse Gas Emissions in Support of National Inventory Reporting to UNFCCC

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Globally, emissions of long-lived non-CO₂ greenhouse gases (GHG; methane, nitrous oxide and halogenated compounds) account for approximately 30 % of the radiative forcing of all anthropogenic GHG emissions. In industrialised countries, 'bottom-up' emission estimates are provided with relatively large uncertainties for non-CO₂ GHGs when compared with those of anthropogenic CO₂. 'Top-down' methods based on the observation of atmospheric concentrations and applied on the country scale offer an independent tool to reduce these uncertainties and detect biases in emissions reported to the UNFCCC. Hence, they provide additional support for policy makers to control the effectiveness of initiated emission reduction measures and build mutual trust between governments.

Since 2012, the Swiss national inventory reporting (NIR) contains an appendix on 'top-down' studies for selected halogenated compound. More recently, this appendix was extended to include methane and nitrous oxide. Here, we present these updated (FOEN, 2020) regional-scale (~300 x 200 km²) atmospheric inversion studies for non-CO₂ GHG emission estimates in Switzerland, making use of observations on the Swiss Plateau (Beromünster tall tower) as well as the neighbouring mountain-top sites Jungfrauoch and Schauinsland.

We report spatially and temporally resolved Swiss emissions for CH₄ (2013-2019), N₂O (2017-2019), and total Swiss emissions for hydrofluorocarbons (HFCs) and SF₆ (2009-2019) based on a Bayesian inversion system and a tracer ratio method. Both approaches make use of transport simulations applying the regional-scale (7 x 7 km²) Lagrangian particle dispersion model (FLEXPART-COSMO). We compare these 'top-down' estimates to the 'bottom-up' results reported by Switzerland to the UNFCCC. Although we find good agreement between the two estimates for some species (CH₄, N₂O, Figure 1), emissions of other compounds (e.g., HFC-134a) show larger discrepancies. Potential reasons for the disagreements are discussed. Currently, our 'top-down' information is only used for comparative purposes and does not directly feed back into the revision of emission factors in the 'bottom-up' inventory. However, as a result of our long and fruitful collaboration with the national inventory group and similar such efforts in other countries (e.g., UK, Australia, New Zealand) the value of "top-down" methods on a country scale is now being accepted by a broader community. As such, "top-down" methods have recently been recommended in the "2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories" (Volume 1, section 6.10.2, Comparisons with atmospheric measurements), which is part of the legal framework of the UNFCCC emission reporting procedure.

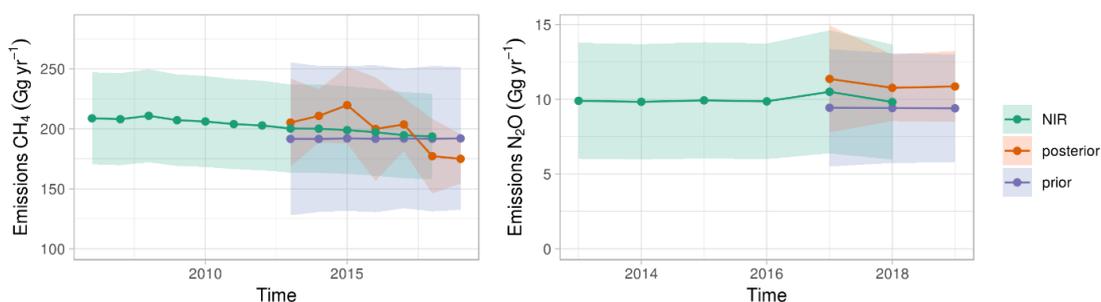


Figure 1. Swiss annual mean anthropogenic emissions of CH₄ (left) and N₂O (right) based on the National Inventory Report (NIR, green line) and based on top-down estimation from atmospheric observations at Beromünster, Jungfrauoch and other sites (posterior, orange line). The shaded areas denote the 1 σ uncertainties ranges. Figures from Swiss NIR report (FOEN, 2020).

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16.6

Metrology for climate relevant volatile organic compounds – MetClimVOC

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Volatile organic compounds (VOCs) play an important role in the atmospheric chemistry, especially in the oxidative capacity of the lower atmosphere (Atkinson & Arey 2003). VOCs are direct and indirect greenhouse gases and precursors of ozone and aerosols, which both contribute to the radiative forcing (Chameides & Walker 1976; Taipale et al. 2008). Accordingly, long-term, accurate and traceable measurements of VOCs are needed to produce comparable datasets at regional and temporal scales, which are essential to identify climate trends.

VOCs are emitted into the atmosphere from natural and anthropogenic sources. Vegetation is the main source of VOCs emitting biogenic VOCs (BVOCs) such as isoprene and monoterpenes (Guenther et al., 1995; Sindelarova et al. 2014). The chemical degradation of BVOCs and the resulting intermediate products are relevant to air quality and climate. A better understanding of BVOC chemistry and quantification of the emissions are essential to predict BVOC feedbacks in the biosphere-atmosphere-climate system. The lack of stable and traceable standards to the international system of unit for some terpenes, together with effects linked to BVOC reactivity with surfaces (i.e. memory effects, decomposition artefacts) and to ozone and humidity interferences are common issues for sites monitoring BVOCs in the atmosphere and BVOC emissions. Similar problems occur for other compounds like oxygenated and halogenated VOCs.

The project “Metrology for Climate Relevant Volatile Organic Compounds” (MetClimVOC, 2020-2023) pursues to minimise these limitations by generating gas standards of oxygenated VOCs, terpenes and halogenated VOCs that are considered priority by stakeholders (WMO-GAW, AGAGE, ACTRIS, EMEP). These gas standards will be produced at atmospheric amount fraction level with an accuracy and temporal stability fulfilling the Data Quality Objectives of the monitoring networks (amount fraction between 1 nmol/mol and 1 µmol/mol with expanded uncertainty < 5% for oxy-VOCs and terpenes and < 1 nmol/mol with expanded uncertainty < 3% for halogenated VOCs; minimum 2 years of temporal stability for gravimetric standards). Moreover, the project aims to optimize the sampling and analytical methods used in monitoring stations and to develop fit-for-purpose working standards.

The project MetClimVOC is part of the European Metrology Programme for Innovation and Research (EMPIR) of the European Association of Metrology Institutes (EURAMET), involves 13 partners from metrological and atmospheric monitoring communities and is coordinated by the Federal Institute of Metrology METAS. Here, we present the project and its first outputs, such as a priority list of VOCs relevant for climate research and their metrological requirements elaborated by active collaboration with stakeholders.

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16.7

The impact of turbulence parameterization in high resolution inverse modelling with FLEXPART-COSMO

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Regional-scale atmospheric inverse modelling can provide observation-based estimates of greenhouse gas emissions at a country scale and, hence, makes valuable information available to policy makers when reviewing emission mitigation strategies and confirming the countries' pledges for emission reduction. Considering that inverse modelling relies on accurate atmospheric transport modelling, any advances to the latter are of key importance. Within the SNSF project IHALOME (Innovation in Halocarbon Measurements and Emission Validation) we characterize and improve the Lagrangian particle dispersion model (LPDM) FLEXPART-COSMO at kilometer-scale resolution and to provide estimates of Swiss halocarbon emissions by integrating newly available halocarbon observations from the Swiss Plateau at the Beromünster tall tower. The transport model is offline coupled with the analysis fields generated by the regional numerical weather prediction model (NWP) COSMO run at MeteoSwiss. Previous inverse modelling results for Swiss greenhouse gases are based on a model resolution of 7 km x 7 km (ref¹). Here, we utilize higher resolution (1 km x 1 km) fields to drive FLEXPART and compare these to previous results.

The higher resolution simulations exhibit increased three-dimensional dispersion, leading to a general underestimation of observed tracer concentrations at the tall tower site Beromünster and when compared to the coarse model results. Because the TKE (Turbulent Kinetic Energy) values do not differ significantly between the two model versions head-to-head comparisons of parameterized turbulence cannot fully explain the concentration discrepancies. Comparisons of variability (turbulence) resolved on the grid-scale suggest that the dispersion differences may originate from a duplication of turbulent transport, on the one hand, covered by the high resolution grid of the Eulerian model and, on the other hand, diagnosed by FLEXPART's turbulence scheme. In an attempt to tune FLEXPART-COSMO's turbulence scheme at high resolution, we scale FLEXPART's parameterized turbulence so it matches the TKE computed in COSMO. Test simulations with the scaled FLEXPART turbulence show remarkable improvements in the high resolution model's ability to predict the observed tracer concentrations at the Beromünster tall tower.

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16.8

Particulate Matter on Honeybee Body Surfaces

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An unconventional method to get an overlook on aerosol particles deposited in the environment is to analyze particles attached on the bodies of honeybees (*Apis mellifera*, L.). During foraging, other particles than pollen accumulate on the bee surface. Based on the chemistry and the morphology of these particles obtained by Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDS), a rough source appointment can be made (Negri et al., 2015).

To get an overview over the particles collected by foraging bees, specimens were sampled around beehives in autumn 2019 at four sites in the Canton of Fribourg with different background settings (rural, urban, close to a highway and close to a shooting range). All particles found at the surfaces are in a size range between 1 and 100 μm . Due to carbon coating and the bee's body composition, organic particles like pollen or carbon particles (soot) are difficult to analyze chemically, but in most cases can be distinguished based on morphology. In addition to pollen and soil-derived (dust) particles, which were always present, the particle population contained site specific components.

Heavy metal rich particles containing Pb, Ba, Cu, Hg, Zn and Sn, typical for the composition of gunshot residue, were found on bees from the beehive close to the shooting range. A second sampling campaign has been undertaken at this site in spring/summer 2020.

The results show that the analysis of particles found on the surface of bees are a quick and convenient tool to monitor the predominant sources of aerosol in the vicinity of beehives.

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16.9

Carbon budget of a drained organic agricultural soil with mineral soil coverage

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The agricultural use of organic soils usually requires drainage that leads to high Greenhouse Gas (GHG) emissions and soil subsidence. A proposed strategy to maintain productivity of these soils is to cover them with mineral soil. To quantify the climatic impact of this practice, the net ecosystem carbon balance (NECB) was determined for a pair of covered (Cov) and uncovered reference (Ref) organic soil. Our experimental field site is located in the Rhine Valley, Switzerland, and the grassland is intensively managed. The NECB was determined for two full years (1.3.2018-29.2.2020) by accounting for all relevant carbon fluxes entering and leaving the soil-vegetation system. For this purpose, the net ecosystem exchange of CO₂ (NEE), CH₄ exchange fluxes, carbon removal by harvest, and carbon import by organic fertilizers were measured. The gas exchange for CO₂ and CH₄ was determined by eddy co-variance (EC). In both years, NEE was positive indicating an upward net flux of CO₂ to the atmosphere. However, the magnitude of CO₂ fluxes differed considerably between the two years for both sites. For the first year, NEE was around 600 g C m⁻² yr⁻¹, about 3 to 4 times higher as in second year. Annual CH₄ emissions were marginal at both sites. Considering the NECB the reference site was found to be a net carbon source with higher emissions in the first year. The NECB of the covered site was similar, indicating a marginal effect of the soil coverage on the NECB in both years. We found the groundwater to be an important driving factor for the observed loss of soil carbon at our site. Based on the two year measurement period, we preliminary conclude that covering organic soils with mineral soil might not be an effective climate mitigation option at the Rüthi site.

16.10

First halogenated greenhouse gas measurements at the Beromünster tall tower in Switzerland and emission estimation

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Synthetic halocarbons reach the atmosphere due to a wide range of anthropogenic activities. Once in the atmosphere, long-lived halocarbons act as strong greenhouse gases, and in addition, chlorinated or brominated halocarbons contribute to stratospheric ozone depletion. To monitor abundances and trends of halocarbons, global measurement networks, such as the Advanced Global Atmospheric Gases Experiment (AGAGE), have been put in place. However, to capture regional pollution sources and to validate country-specific bottom-up emission estimates by atmospheric observations, additional measurements, focusing on smaller scales, are required. For this purpose, we already carried out devoted multi-month field campaigns in the Eastern Mediterranean and Eastern Europe. However, halocarbon emissions from many European regions remain largely unexplored in terms of atmospheric observations.

In the frame of the SNF-project IHALOME we present the first continuous halocarbon measurements covering the most industrialized and densely populated area of Switzerland, the Swiss Plateau. The data obtained during a one-year measurement campaign at the Beromünster tall tower in the canton of Lucerne complement the long-term measurements at the high altitude research station Jungfraujoch. High precision, high accuracy measurements of >60 halogenated substances were performed via the analytical setup of the AGAGE network (Miller et al. 2008): Air samples are pre-concentrated in a two-trap process at low temperatures before the analytes are separated by gas chromatography (GC) and detected by quadrupole mass spectrometry (MS). All halogens compound classes of the Montreal and Kyoto Protocols are represented in our measurements, the chlorofluorocarbons (CFCs), the hydrochlorofluorocarbons (HCFCs), the hydrochfluorocarbons (HFCs) and perfluorocarbons (PFCs) and the youngest generation of unregulated hydrochfluoroolefins (HFOs). The obtained results improve our understanding of halocarbon abundances and source allocation in Switzerland, and, for the first time, they offer the possibility to robustly quantify Swiss national halocarbon emissions with top-down methods (tracer ratio and Bayesian inverse modeling).

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16.11

Simulating the effect of decadal soil warming on CO₂ losses from a temperate forests soil

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As a consequence of the predicted warming of the atmosphere over the coming decades, soils are expected to warm at similar rates. This warming might have profound consequences for the rate at which biogeochemical processes take place in soils, such as an enhanced rate of soil organic carbon mineralization to CO₂. Making reliable predictions of the response of soil organic carbon to soil warming is therefore necessary to assess the extent to which this process might constitute a positive feedback to global climatic change over the coming decades.

Therefore, we applied a state-of-the-art soil biogeochemical model (ReSOM, Tang and Riley (2015)) to two long-term soil warming experiments (+ 5 °C) at Harvard Forest (Massachusetts, USA), which have been running for 17 and 29 years. Our aims are to (1) assess the extent to which ReSOM is able to correctly simulate the effect of long-term soil warming on soil organic carbon stocks and temporal patterns of CO₂ fluxes, (2) assess the importance of including thermal adaptation of soil microbes (using macromolecular rate theory) and temporal patterns in soil moisture content in ReSOM to correctly simulate intra- and inter-annual soil organic carbon dynamics, and (3) check the reliability of model predictions to make forecasts on decadal timescales when calibrated using data from a decade of soil warming.

Our results show that including thermal adaptation of soil microbes is necessary to reliably predict the effect of decadal soil warming on intra- and inter-annual CO₂ fluxes. In addition, the limiting effect of soil moisture on microbial decomposition of soil organic carbon needs to be simulated in order to correctly simulate the effect of soil warming at different forest sites in the same region. Last, we assessed the effect of including or excluding the effect of (1) soil moisture and (2) thermal adaptation of soil microbes on simulated changes in soil organic carbon stocks over the next century. We found diverging predictions of soil organic carbon losses under these different scenarios, showing the importance of correctly including and parameterizing these processes.

REFERENCES

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16.12

The impact of mineral soil coverage on N₂O emissions from organic soil drained for agriculture

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Peatland store 12 – 21 % of the total soil organic nitrogen (N), which accumulated over millennia. However, long - term drainage of peatland for agricultural use leads to a strong release of carbon and nitrogen and subsidence of peatland through aerobic peat decomposition. In order to improve the sustainability of peatland management in agriculture, and to counteract soil subsidence, mineral soil coverage is becoming an increasingly used practice in Switzerland. Mineral soil coverage may affect the N balance from the corresponding organic soil, owing to the eventual change of surface soil characteristics. However, the effect of mineral soil coverage on nitrous oxide (N₂O) emission has not been studied yet. Here, we report on a field experiment carried out to explore the impact of mineral soil coverage on the N₂O emission from drained organic soil.

The experimental site, a drained peatland with a peat thickness of around 10 m, is located in the Swiss Rhine Valley. In 1973, an integral drainage system was built. Since then, an intensively managed meadow was established, with mineral and slurry fertilization and 5 to 6 grass cuts per year. In 2006, one part of the field (1.7 ha) was covered with mineral soil material (thickness 30 – 40 cm). We established our field experiment on this mineral soil coverage site (DC) and used the adjacent drained organic soil without mineral soil coverage as reference (DN). Both sites have the identical management and vegetation. In our experiment, an automatic chamber system is used for collecting the N₂O at an interval of 3 h. After one and a half year's (03.2019 to 08.2020) continuous measurement, the data reveals that: the average N₂O emissions from DN (10.66 ± 1.28 mg N₂O-N m⁻² day⁻¹) exceeds the one from DC (1.14 ± 0.08 mg N₂O-N m⁻² day⁻¹) by a factor of 10. The more details analysis shows that this difference between DC and DN is mainly driven by the different reaction to fertilizer inputs. In general, the N₂O peaks occur shortly after N application and last for 2 to 3 weeks before returning to background emission. The N₂O peaks after fertilization account for 80 % and 70 % of the total N₂O emissions for DN and DC, respectively. However, significantly higher peak N₂O emissions were found in DN than DC, whereas the background N₂O emissions show no difference. To further explore the impact of mineral soil coverage on the N balance of drained organic soil, extra ¹⁵NH₄¹⁵NO₃ labelled fertilizer will be applied in September 2020 to trace the N transformation at DC and DN. In summary, our data suggest that mineral soil coverage could strongly reduce N₂O emission from drained organic soil, and may therefore be an interesting GHG mitigation measure in agriculture.

