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Validation and application of Open-Path FTIR in combination with a micrometeorological transport model for measuring CH_4, NH_3, and N_2O fluxes from open digestate storage lagoons

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Biogas production for generating electricity is explicitly promoted in several European countries as a regenerative and greenhouse gas (GHG) saving energy source. However, also during biogas production significant GHG (especially methane, CH_4, and nitrous oxide, N_2O) and ammonia (NH_3) emissions occur and reduce not only GHG savings from biogas energy but also pose additional threats to natural ecosystems by nitrogen deposition. Anaerobic digestates (AD), the residues of the biogas fermentation process, are usually stored in large tanks or lagoons for several months prior to land spreading as organic fertilizers. When uncovered, these stores pose high potential for NH_3 and GHG emissions. However, measurements of these emissions are methodically challenging. In the present study the use of Open-Path Fourier Transform infrared spectroscopy (FTIR) as a non-invasive remote sensing approach for determining CH_4, NH_3, and N_2O emissions from two open AD storage lagoons with a surface of 1100 – 1200 m^2 in combination with a backward Lagrangian Stochastic dispersion technique (bLS) was validated and deployed.

CH_4, NH_3, and N_2O concentrations were measured on a 30 to 50 m optical path by OP FTIR, while wind speed and direction were monitored by a sonic anemometer. Air temperature and pressure were logged as well. Three gas release experiments were conducted, in which N_2O or NH_3, respectively, was released from a grid at the lagoon surface, and the gas recovery rate of the bLS technique was determined for validation of the applicability of the combination of FTIR and bLS under local conditions not optimal for the application of micrometeorological techniques. In these experiments the ratio between calculated (Q_{bLS}) and actual flux (Q) was 0.89 ($\pm$0.04; n = 24), 1.14 ($\pm$0.13; n = 23), and 1.23 ($\pm$0.1; n = 59). Over all three release experiments the (Q_{bLS}/Q) ratio was 1.13 ($\pm$0.17; n = 106).

Trace gas emissions from both lagoons were dominated by CH_4, which was found up to 47.6 kg CH_4 C day^{-1} from one of the lagoons. NH_3 emissions were up to 4.9 kg NH_3 N day^{-1}. N_2O emissions were only detectable during summer with up to 0.7 kg N_2O N day^{-1}. Destruction of a naturally occurring surface crust by mechanical homogenization resulted in immediate high CH_4 peaks, but did not affect total CH_4 release. In contrast, NH_3 emissions were raised by up to six times and remained elevated for the following days, while N_2O fluxes increased by about 35%. CO_2 equivalent weighted emissions from the lagoons per unit electricity produced by the corresponding biogas plant ranged between 7 and 218% (half-fermented AD pumped into lagoon due to problems in fermenter) of the CO_2 emissions arising from utilization of equal amounts of natural gas, depending probably on factors like season, biogas plant, and lagoon filling level. A full assessment of GHG emissions from biogas production would require a more systematic investigation. Nevertheless, the high trace gas emissions during AD storage clearly counteract the purpose of biogas energy, which is mainly motivated by a reduction of GHG emissions. Gas tight covers on AD storage facilities are strongly recommended to reduce these GHG emissions and to raise the efficiency of biogas energy.