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Simultaneous Measurement of NH₃, N₂O and CH₄ to Assess Efficiency of Trace Gas Emission Abatement After Slurry Application

By

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S u m m a r y

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The application of liquid organic fertilizers can directly or indirectly induce the emission of trace gases. There are already a number of investigations on mitigation strategies, but most of them focus on one gas only and neglect the fact, that conditions reducing the emission of one trace gas might have a directly opposed effect on another.

We conducted several field experiments to investigate the influence of application technique and pre-treatment of slurry on trace gas emissions after slurry application. Ammonia, CH₄ and N₂O were measured simultaneously. It could be clearly shown, that some of the tested slurry management techniques had contrary effects on the emissions of the different trace gases. With information on emissions of single trace gases, wrong conclusions might be drawn concerning the effectiveness of mitigation options.

By comparing the emissions based on CO₂ equivalents, the different management options can be compared comprising all three studied trace gases.

I n t r o d u c t i o n

Organic waste from animal livestock is a major source for trace gases. During storage and after application methane (CH₄), nitrous oxide (N₂O) and ammonia (NH₃) are emitted.

The interactions of slurry, soil and atmosphere controlling the emissions after application are very complex. In this system soil can be described as a physi-

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cal boundary and a microbiological reactor. Slurry provides nutrients and metabolizable organic matter for microbial activity and is subjected to physico-chemical processes leading to emission of dissolved gases. Viscosity of the slurry strongly influences, how fast it infiltrates into the soil.

In order to reduce emissions following slurry application, different application techniques and slurry treatments have been investigated concerning their impact on trace gas emissions.

Ammonia emissions may be reduced effectively by using application techniques that incorporate slurry into the soil. But FLESSA & BEESE 2000 found that slurry injection may enhance N_2O emissions. Anaerobic digestion can reduce N_2O emissions after slurry application on the one hand (PETERSEN 1999). On the other hand there might be a potential for higher NH_3 emissions due to a higher pH in the digested substrates (KTBL 1998). Other treatments may be stripping of NH_3 (HAHNE 1996) or slurry acidification (BUSSNIK & al. 1994).

Most field studies on emissions of trace gases focused either on the direct greenhouse gases CH_4 and N_2O , or on NH_3 that contributes to eutrophication and is an indirect greenhouse gas. Only few studies exist on the overall impact of slurry application on greenhouse gas emissions considering both direct and indirect greenhouse gases (REITZ & SCHÜRER 1999, SCHÜRER 2000, WESLIEN & al. 1998). In this paper we summarize results of three field studies conducted in 1998 and 1999 which were integrated in different projects, but had a common experimental design. We measured NH_3 , N_2O , and CH_4 with various slurry treatments and application techniques and calculated the overall emissions in terms of CO_2 equivalents. As processes determining emissions of these trace gases are already described in various studies, no courses of emission rates will be shown, but cumulative emissions only, allowing to assess the effectiveness of different mitigation options after slurry application.

Materials and Methods

Slurry treatments

No treatment

Untreated beef cattle slurry.

This slurry was taken from the same farm as the anaerobically digested slurry, and was processed further to provide the substrate for the acidified treatment (see below).

Anaerobic digestion

The product of a biogas plant fermenting the cattle slurry mentioned above was used. Mean duration of fermentation was 28 days at 37-40°C. Slurry and the fermentation product differed in their chemical parameters (Table 1). During fermentation organic compounds are digested by methanogenic microorganisms producing CO_2 and CH_4 . Anaerobic digestion reduces slurry dry matter content and easily degradable carbon measured as biological oxygen demand within 5 days (BOD_5). The increase of pH and NH_4^+ content was not as high as expected from other investigations. Reason might be, that digestion was performed on farm in a flow through system and not a batch reactor. Therefore it was not possible to sample exactly the raw slurry from which the digested slurry originated.

Fermentative acidification

The addition of microbial available carbon is described in a review on additives to reduce ammonia emissions by MCCRORY & HOBBS 2001. Sugar is an expensive but well defined labile carbon source. But other materials such as milled wheat may be used as well for an internal acidification (own experiments). For demonstration purposes, sugar was used in this experiment.

1kg of sugar was added to 50 litre of cattle slurry. Microbial production of organic acids temporarily decreased the pH significantly. Slurry was applied 2.5 days after sugar addition, when pH 6 was reached. The lower $\text{NH}_4\text{-N}$ content in the acidified slurry compared to the untreated slurry (Table 1) may be due to a delayed sugar addition (24 hours). During this period the vessel allowed gas exchange and water evaporation. After sugar addition, the high microbial activity may have led to a higher microbial biomass going along with a higher total N content compared to untreated slurry.

Co-fermentation

This substrate was produced through combined anaerobic fermentation of 50 % cattle slurry together with 50 % biowaste (organic household waste) in a biogas-plant. Mean duration of fermentation was approximately 40 days under mesophile temperature conditions (42°C). The changes in chemical and physical properties depend on the co-substrate but are in general similar to the changes described for anaerobic digestion of slurry alone, resulting in higher pH and $\text{NH}_4\text{-N}$ content, but lower dry matter content than undigested slurry.

Table 1. Parameters of slurry treatments.

	slurry	anaerobically digested slurry	fermentative acidified slurry	co- fermentation
pH	6.7	6.8	6.0	8.9
dry matter [%]	9.2	5.1	10.0	4.8
$\text{NH}_4\text{-N}$ [g/kg]	1.6	1.5	1.2	2.2
total N [g/kg]	3.7	3.2	3.8	3.8
total C [g/kg]	36.8	19.5	40.0	n.d.
BOD_5 gO_2/kg	13.9	6.9	14.4	3.0

Application techniques

A co-fermentation product was used in these experiments, because they were integrated in a project on the evaluation of trace gas emissions through co-fermentation. Due to the small plots (9 m²) the different application techniques were simulated by hand, apart from injection where a custom made tractor drawn device was used. 30 m³ ha⁻¹ were added to the plots using watering cans, modified to simulate splash plates, trailing hoses and trailing shoes. On arable land one of the trailing shoe treatments was immediately shallow incorporated using a garden harrow. Application of substrate on a single plot took about 10 minutes and gas flux measurement started immediately thereafter. The substrate was applied to the all plots within 90 minutes.

Experimental setup

Application technique experiments

The experiments were conducted on grassland and farmland in the western part of Germany near the city of Bonn. The grassland experiment (Heidgen) was started in April 1999 on a poorly drained Stagno-gleyic Luvisol (FAO) with rather wet and cool weather at the beginning of the study. Water filled porosity decreased from 90 % to 60 % during the first two weeks. Mean daily temperature decreased from 8 °C to 2 °C and night frost after 7 days. From the second week until the end of the study after 6 weeks, mean daily temperatures fluctuated between 10 and 15 °C. The trial on arable land (Klein-Altendorf) was started three weeks later on a well drained Luvisol when the rather warm and dry weather prevailed.

The two experiments were both carried out in a completely randomized design with 4 replications for each treatment. Plot size was 9 m² with 10 m spacing between the plots. CH₄ and N₂O emissions were measured after application of 30 m³ ha⁻¹ of co-fermentation product, with very intense sampling every 6 hours during the first four days. Measurements were then performed daily for two weeks, later once per week until 6 weeks after application. NH₃ measurements were conducted during the first four days after spreading only.

Slurry treatment experiment

Soil type of this experiment was a Luvisol with loamy texture in the city of Bonn. Experimental design was similar to the application experiments, but plot size was 4 m² with 3 to 5 m spacing between the plots and only 20 m³ ha⁻¹ of slurry was added. Measurements of NH₃ volatilization were carried out during three days after application in Mai 1998. CH₄ emissions were measured during the first 36 hours and N₂O emissions for 6 weeks, sampling every 12 hours during the first three days. The following 2 weeks N₂O samples were taken every second day and for the rest of the experiment every fourth day.

Gas measurements

NH₃ volatilization

Ammonia volatilization was measured using the standard comparison method described by VANDRÉ & KAUPENJOHANN 1998. This is an open method where ammonia is collected in passive samplers installed on each plot 15 cm above the soil surface. Passive samplers were filled with 0.05 M H₂SO₄ and the acid was changed every 4 to 6 hours. Ammonium concentrations in the passive sampler solution thus gave mean signals over this period and were analysed colometrically using a continuous flow analyser (Bran & Lübbe Autoanalyser). On standard plots of the same size and exposition as the experimental plots, known amounts of NH₃ were emitted. In the substrate-experiment this was achieved with plastic channels containing slurry that was tested for NH₄-N loss during each sampling period. In the application experiment a more convenient design was used, which released NH₃ from gas cylinders through a tube system. In the studies conducted by VANDRÉ & KAUPENJOHANN 1998 these methods agreed rather well with each other. Knowing the flux rate from the standard plots and the ammonium concentration in the passive sampler solutions collected from these plots, a transfer factor for each sampling period could be determined and allowed the calculation of flux rates on the experimental plots from passive sampler ammonia concentrations.

N₂O and CH₄ emission

Nitrous oxide emissions were determined using closed chambers (HUTCHINSON & MOSIER 1981) covering a surface area of 0.25 m² with a volume of 96 dm³ in the application experiment and 0.14 m² with 85 dm³ in the slurry treatment experiment. Gas samples were taken from the chambers with evacuated headspace vials (0.02 dm³) through a butyl septum. Samples were taken 0, 0.5, 1, and 1.5 hours after placing the chambers air-tight onto installation rings that were permanently inserted into the soil surface. Gas analysis was performed using a gas chromatograph

(SRI 8610C) with electron capture detector (ECD) for nitrous oxide and flame ionization detector (FID) for methane.

Results and Discussion

NH_3 volatilization

Ammonia emissions were highest immediately after application (data not shown). As already described in various studies (e.g. WESLIEN & al. 1998, SOMMER & JACOBSEN 1999), most of the emissions occurred during the first 24 hours and volatilization after 3 days was neglectable.

The influence of application technique and substrate treatment on NH_3 volatilization can clearly be shown with cumulated emissions. In order to compare the emission from the different experiments the losses are given in % of added $\text{NH}_4\text{-N}$, because different amounts of slurry were used for fertilization in the substrate and in the application experiments (Fig. 1).

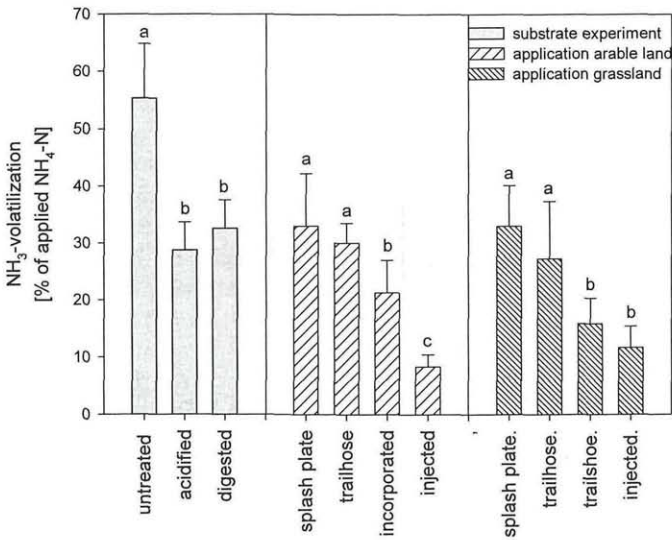


Fig. 1. NH_3 volatilization from the different treatments. Expressed as percentage of applied $\text{NH}_4\text{-N}$. Means and standard deviation ($n=4$). Different letters indicate significant differences within individual experiments ($p<0.05$).

Emissions after digestion or acidification of the slurry were significantly lower ($p<0.05$) compared to untreated slurry. The causes of reduced NH_3 losses from digested or acidified slurries have very different reasons. Acidification decreased the pH of the substrate retarding NH_3 volatilization, whereas digestion re-

sulted in smaller dry matter content and thus allowed faster infiltration of the substrate. This confirms experiments conducted by RUBAEK & al. 1996 who report of NH_3 emissions of approximately 35 % after trail hose application of digested slurry compared to 45 % from untreated slurry. Nevertheless this reduction of NH_3 losses seems to be limited to warm and dry weather, because in the same study, when infiltration was generally impeded due to high soil moisture content, no reduction of NH_3 emissions could be observed.

Trailhose application of co-fermentation product in the application experiment resulted in similar percentages of NH_3 losses as from trailhose applied anaerobically digested slurry in the substrate experiment (Fig. 1). Direct comparison of these experiments is not possible, as site characteristic and amount of slurry added were different. However it seems that dry matter reduction through digestion is a very important factor for the reduction of ammonia emissions. Apart from digesting the slurry, further reduction of NH_3 emissions was achieved by using appropriate application techniques. The more the substrate was incorporated into the soil or applied near the soil on the grassland site, the less NH_3 was lost. Injection of the substrate reduced losses to less than 10 % of the applied $\text{NH}_4\text{-N}$ on both sites whereas losses after splash plate application amounted to about 30 %. Spreading with trail hoses resulted in almost the same NH_3 volatilization as splash plate on the arable site, because the fermented substrate was of low viscosity having small dry matter content and could not be applied in defined bands but spreaded over a greater surface area due to encrustation of the bare soil surface. On the grassland site, trail hose application performed slightly better (not significant), as some of the substrate filtrated through the grass and thus was less exposed to atmosphere and wind. When the substrate was applied directly to the soil surface with trailing shoe on the grassland site, emissions were as low as after injection, most probably because resistance of diffusion through the grass was high and the substrate could infiltrate directly into the soil. On the arable site, shallow incorporation immediately after trailhose application substantially reduced emissions, but was less efficient than injecting the substrate. Magnitude and pattern of reduction in NH_3 emissions are similar to results of other studies on incorporation or injection of slurry (REITZ & al. 1999, VAN DER MOLEN & al. 1990, DOSCH & GUTSER 1996). It can be concluded, that the more the substrate is incorporated into the soil, the less NH_3 is lost. Nevertheless it should be considered, that overall emissions of NH_3 also strongly depend on climatic conditions (VANDRÉ & al. 1997, SOMMER & al. 1991).

CH_4 emission

Methane emissions from all surface applied substrates were high immediately after application and hardly detectable during following measurements (data not shown, see also CHADWICK & al. 2000). This indicates, that dissolved methane in the substrates might be the only source of emissions and that no production of methane after application took place. Very high emissions immediately after application, probably even before measurements could be started, result in a high uncertainty concerning cumulated emissions. Therefore it is difficult to compare treat-

ments with different time courses of emissions. Especially emissions after injection differed from the other application techniques, with emissions continuing for 24 hours after application, most probably due to physical restraint of CH_4 diffusion. In the few studies on methane emissions after field application of slurry cumulated emissions differ considerably. CHADWICK & al. 2000 describe two experiments. Under rather dry and warm conditions with only brief and low methane emissions immediately after application, the soil acted as a minor sink for CH_4 in the 3 weeks after application. When wetter and colder climatic conditions prevailed, emissions amounted to 580 g CH_4 within 3 weeks. Also SCHÜRER 2000 and REITZ & al. 1999 report of comparably high CH_4 emissions with 1200 g to 2900 g ha^{-1} within one day.

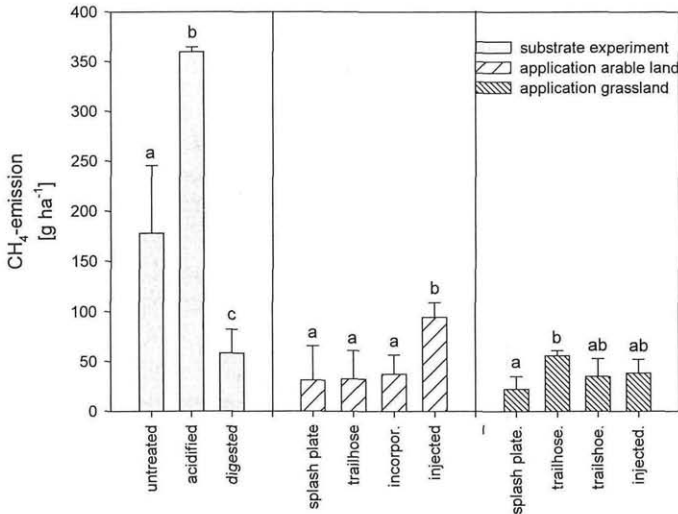


Fig. 2. Cumulated CH_4 -emissions after application of differently treated slurries and co-fermentation product applied with different application techniques. Means and standard deviation ($n=4$). Different letters indicate significant differences within individual experiments ($p < 0.05$).

Comparing cumulated emissions from the treatments it has to be taken into account that in the substrate experiment 20 m^3 and in the application experiment 30 m^3 of slurry were applied. Nevertheless it seems, that application technique only has a minor effect, whereas slurry treatment has greater impact (Fig. 2). Especially anaerobic digestion of the slurry reduced methane emissions after field application, most probably due to the fact that easily degradable organic compounds were already converted to CO_2 and CH_4 during the digestion in the biogas plant, as can be confirmed by smaller BOD_5 contents in both digested substrates: slurry with and without co-fermentation. Thus prior to application less methane is produced and dissolved in these substrates.

After application of acidified slurry, highest amounts of methane were emitted. Fermentative acidification of the slurry was achieved by microbial production of organic acids after sugar addition. These acids are easily degradable by micro-organisms and might to some extent be converted to CH_4 and CO_2 during storage of the substrate, resulting in higher amounts of dissolved CH_4 . However it is known that methanogene bacteria are pH-sensitive and at a pH of 6 CH_4 formation is depressed (SCHLEGEL 1985). The high amounts of CH_4 emitted after field spreading may have been formed during the fermentation process before a low pH was reached and emission was suppressed until field application. Since no data are available on CH_4 formation during the fermentative process this remains speculative.

N_2O emission

The time course of N_2O emissions was different from those of NH_3 and CH_4 . As described already from other authors (FERM & al. 1999, MOGGE & al. 1999) emission rates were not only high immediately after slurry application, but further emission peaks occurred when environmental conditions like soil water content and temperature were favourable. Different emissions from the control plots reflect differences in environmental conditions during the three experiments (Fig. 3). Especially in the substrate experiment high background emissions of nitrous oxide took place.

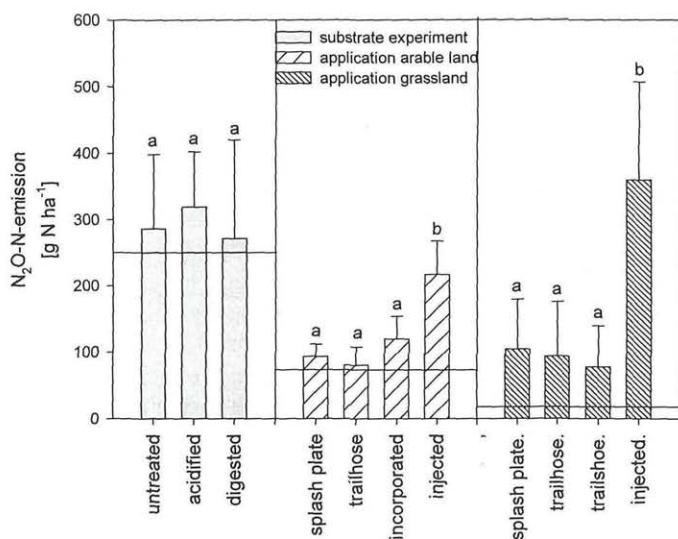


Fig. 3. Cumulated $\text{N}_2\text{O-N}$ emissions during 5 weeks after application of different slurries and co-fermentation product applied with different application techniques. Means and standard deviation ($n=4$). Horizontal lines indicate levels of background emissions from unfertilized plots. Different letters indicate significant differences within individual experiments ($p < 0.05$).

Cumulated slurry induced nitrous oxide emissions were not significantly different for the three substrate treatments (Fig. 3). Only during a burst of emissions immediately after application, emission rates were different ($p > 0.05$) with 322 and 258 $\mu\text{g N}_2\text{O-N m}^{-2} \text{ h}^{-1}$ for raw slurry and acidified slurry compared to 58 $\mu\text{g N}_2\text{O-N m}^{-2} \text{ h}^{-1}$ after spreading digested slurry (data not shown). The much lower content of easily degradable carbon (BOD_5) in the digested slurry might be the reason for this much smaller emission peak. This is in line with results from other studies with C-reduced slurries conducted with fermented (PETERSEN 1999, RUBAEK & al. 1996) or separated slurries (DOSCH & GUTSER 1996). High background emissions of nitrous oxide and high variability of emissions during the remaining 6 weeks of the experiment might be the reason for the only small and not significant reduction in emissions through slurry digestion in our experiment. Comparing different application techniques, significantly more nitrous oxide emitted after injection of the slurry, but no difference could be found between the other application treatments. Elevated emissions after injection might be due to retarded oxygen diffusion and higher water content in greater depth of the soil promoting denitrification processes.

Global warming potential (GWP)

The emissions of the different trace gases are affected in different manners by the slurry treatments and application techniques. In our experiments, nitrous oxide emissions were only little and not significantly influenced by slurry treatment, whereas both acidification and anaerobic digestion had a considerable effect on NH_3 and CH_4 emissions. Injection of the substrate effectively reduced NH_3 volatilization, but increased N_2O emissions two- to three-fold. In order to evaluate the potential of the tested slurry treatments and application techniques to reduce greenhouse gas emissions as a whole, emissions of the different trace gases were converted into CO_2 equivalents using IPCC conversion factors (IPCC 1996). These factors are calculated from absorption efficiency of long-wave radiation and mean residence time in the atmosphere. They amount to 310 for N_2O and 21 for CH_4 .

Ammonia is not considered a direct greenhouse gas because of its short lifetime in the atmosphere, but its immission induces N_2O formation elsewhere. It is postulated, that 1 % of emitted $\text{NH}_3\text{-N}$ is converted to $\text{N}_2\text{O-N}$ (IPCC 1997). Therefore it is possible to integrate NH_3 losses into GWP calculations. Nevertheless it should be taken into account, that detrimental effects of NH_3 also include acidification and eutrophication. These effects are not subject of this paper, but should be taken into account, when choosing certain slurry treatments or application techniques.

In order to compare slurry induced emissions only, emissions from the control plots (no fertilization) were subtracted from overall emissions.

The contribution of NH_3 and N_2O emissions to global warming is evident, whereas CH_4 emissions after field application seem to be of minor relevance (Fig. 4). This is in line with CHADWICK & al. 2000, who report that CH_4 produced by enteric fermentation in ruminants is much more important. Measures designed to

reduce ammonia emissions, such as slurry treatment and application techniques, can be very efficient, but bear the risk of increased N_2O emissions.

When emissions of CO_2 equivalents from the different treatments in our experiments are compared, digestion of slurry and trailshoe application on grassland can be identified as effective measures to reduce greenhouse gas emissions.

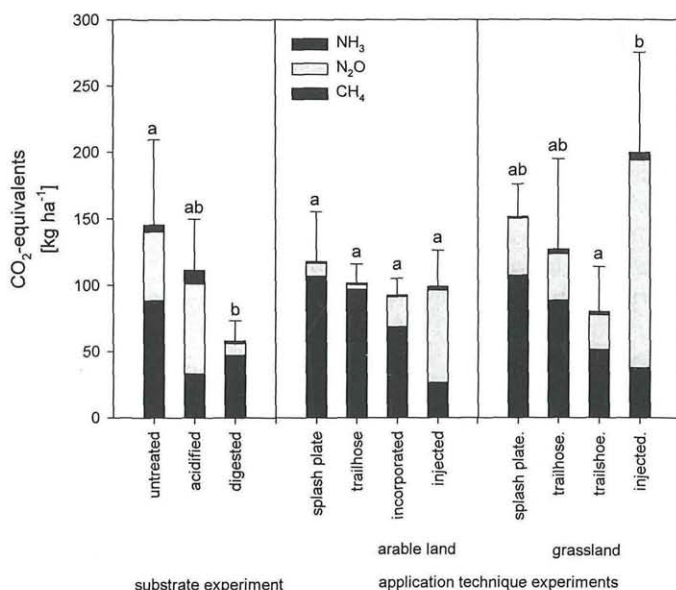


Fig. 4. CO_2 equivalents calculated from the cumulated emissions of the different trace gases. Means ($n=4$). Error bars are calculated for the sum of equivalents. Different letters indicate significant differences within individual experiments ($p < 0.05$).

Conclusions

The emissions of the different trace gases are influenced by very different factors. These are of physical nature for NH_3 and microbial nature for N_2O . Methane emissions seem to depend mainly on CH_4 production prior to application. The effect of management practices on these trace gases can be integrated by calculating CO_2 -equivalents. Our results implicate, that digestion of slurry has a positive overall effect on trace gas emissions. The emissions of all monitored trace gases were reduced compared to unfermented slurry, although the effect on nitrous oxide was not clear in our experiment. For spreading co-fermented slurry on grassland trail shoe application seems to be the most recommendable, especially if the effect of ammonia on eutrophication and acidification is considered. On arable land the results were not as clear, but it should be considered, that in addition to the mentioned biological sources of trace gases, injection of slurry causes higher fuel con-

sumption with negative effects on greenhouse gas budgets. In order to assess environmental effects and economic feasibility of slurry management practices, further studies are needed, that integrate additional aspects such as applicability to certain crops, site exposition, soil structure and cost of machinery.

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