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Nitrous oxide emission reduction in temperate biochar-amended soils

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Abstract

Biochar, a pyrolysis product of organic residues, is an amendment for agricultural soils to improve soil fertility, sequester CO_2 and reduce greenhouse gas (GHG) emissions. In highly weathered tropical soils laboratory incubations of soil-biochar mixtures revealed substantial reductions for nitrous oxide (N_2O) and carbon dioxide (CO_2). In contrast, evidence is scarce for temperate soils. In a three-factorial laboratory incubation experiment two different temperate agricultural soils were amended with green waste and coffee grounds biochar. N_2O and CO_2 emissions were measured at the beginning and end of a three month incubation. The experiments were conducted under three different conditions (no additional nutrients, glucose addition, and nitrate and glucose addition) representing different field conditions. We found mean N_2O emission reductions of 60 % compared to soils without addition of biochar. The reduction depended on biochar type and soil type as well as on the age of the samples. CO_2 emissions were slightly reduced, too. NO_3^- but not NH_4^+ concentrations were significantly reduced shortly after biochar incorporation. Despite the highly significant suppression of N_2O emissions biochar effects should not be transferred one-to-one to field conditions but need to be tested accordingly.

1 Introduction

In future mankind has to cope with global warming as well as with shortage of agricultural land for food production (IPCC, 2007). Application of mineral fertilizer and animal manure has increased agricultural productivity but provoked, among other environmental harms, an increase in nitrous oxide (N_2O) emissions (Denman et al., 2007). N_2O has a 300 times higher global warming potential than CO_2 and is currently the dominant source of ozone depleting nitrogen oxides in the stratosphere, thus having a dual negative impact on the environment (Ravishankara et al., 2009). Thus measures to reduce N_2O emissions from agriculture while at the same time restoring or enhancing agricultural productivity are called for.

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Biochar application to soil is seen as a strategy to improve soil productivity, sequester atmospheric CO_2 (long term sink, Lehmann et al., 2006) and mitigate N_2O emissions at the same time. Biochar is pyrolysed biomass from various organic feedstock which is produced by heating in an oxygen-limited environment (pyrolysis) at temperatures of 400–900 °C (Lehmann, 2007). There is increasing evidence from laboratory studies that biochar addition positively effects net gas exchange with a 50–80 % reduction for N_2O (Yanai et al., 2007; Spokas and Reicosky, 2009; Spokas et al., 2009; Van Zwieten et al., 2010; Singh et al., 2010; Kammann et al., 2011). Most of the studies so far were conducted in tropical systems and there is a lack of experiments for temperate agricultural soils. Prior to experiments at field scale it is important to test the effects of biochar application on N_2O emissions in the laboratory first.

The mechanisms for reduced N_2O emissions from soil amended with biochar are still in debate. In this study the following three hypotheses are addressed:

1. Reduced nitrogen availability for nitrification and denitrification processes due to adsorption of ammonium (NH_4^+) (Tsutomu et al., 2004; Singh et al., 2010) or nitrate (NO_3^-) (Mizuta et al., 2004) on biochar surfaces.

2. Limited substrate availability through absorption of labile C at biochar surface. Biochar increases the stability of soil carbon (Clough and Condron, 2010) and hence lowers the available substrate for micro-organisms.

3. Alkaline biochars increase the pH of soils which may promote complete reduction of N_2O to N_2 in the denitrification cascade (Stevens et al., 1998).

An important point regarding the sustainability of biochar application to soil is the evolution of GHG fluxes over time. Whereas adsorption capacity of the amended biochar may decrease with time, effects on pH or changes in denitrification rates may remain constant or even increase with biochar maturation (Major et al., 2010).

The main objective of this study was to asses if temperate nutrient rich agricultural soils show reductions in N_2O and CO_2 emissions after biochar application similar to

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tropical soils and how the effect of reduction evolves with ageing of the biochar in the sample. In a three-factorial experiment with two different biochars applied to two intensively managed agricultural soils, the effect of biochar amendment on N_2O and CO_2 emission rates was assessed in laboratory incubations over three months. To stimulate microbial activity experiments were conducted under three different conditions (no additional substrate, glucose addition, nitrate and glucose amendment).

2 Material and methods

Two different *biochars* distributed by the Delinat-Institute, a research association for organic viticulture in Switzerland (Delinat-Institute, 2011) were chosen for the experiment. The biochars were produced in a commercial pyrolysis reactor from Pyreg (2011). The feedstock of the two chars were green waste (gw) and coffee grounds (cg). The maximum temperatures at pyrolysis from estimations by the facility manager are indicated in Table 1. Residence time in the reactor was 25 min. The biochars were analysed for specific surface area (measured by N_2 adsorption of dried (105 °C for 12 h) and sieved (< 2 mm) samples using a Quantachrome Nova 2200 surface analyzer, Odelzhausen, Germany), C- and N-content (elemental content analysed from combusted samples using a Hekatech Euro EA 3000 elemental analyzer, Wegberg, Germany), and pH (measured in 1 : 5 biochar : $CaCl_2$-solution (0.01 M) by ExStik PH100 pH Meter, EXTECH). The properties of biochar and further analysis of elemental composition are shown in Tables 1 and 2, respectively. For homogenisation the biochar was sieved (< 2 mm) before soil addition.

The *soils* used stem from two different temperate agricultural sites in Switzerland, Oensingen (Oe) and Reckenholz (Rh). Table 3 summarises main properties of these soils. For elemental analysis, soils were fumigated in a desiccator with concentrated HCl to remove carbonates. Since 2002 the Oensingen field (7.7° E, 47.3° N at 450 m a.s.l.) was used as an intensively managed grassland site receiving 230 kg N ha^{-1} yr^{-1} and was typically cut 4–5 times a year. The Reckenholz site (8.5° E, 47.4° N at 443 m a.s.l.) was used in mixed rotation with annuals for the last years (2007–2010: mirasol,

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winter wheat, silage maize, winter wheat). The site was fertilized by slurry. Depending on the crop 30–100 kg N ha^{-1} yr^{-1} were applied.

Ten litres soil from the Oensingen site were collected at the 26 October 2010 and around 12 l soil from the Reckenholz site at the 18 October 2010 (both sampled at 0–20 cm depth). The soil was sieved (< 6.3 mm) and plant material and stones were removed manually. Moisture content and dry weight were measured by drying subsamples at 105 °C for 12 h. For the mixture of soil and biochar 2 kg of each soil were mixed with 2 % w/w biochar (dry weight per dry weight). This corresponds to 8 t ha^{-1} which was found a practicable amount for field applications and showed effects on GHG emissions in other studies (e.g., Yanai et al., 2007; Van Zwieten et al., 2010).

The maximum water holding capacity (max. WHC) for all combinations were measured by saturating 40 g of each moist sample with water in a 50 ml glass cylinder with a water permeable and wetted membrane at the lower end. After one hour of infiltration some water was added from the top before letting the samples drip for two hours. Samples were then weighted again. The pH of all samples was measured in a 1 : 2.5 moist soil : $CaCl_2$-solution (0.01 M) using an ExStik PH100 pH Meter, EXTECH. Table 4 lists sample's labels and shows calculated specific surface areas after mixing with biochar, max. WHC, and pH of all samples used for the incubations.

Soil *samples* were filled in steel cylinders of 55 mm diameter, 42 mm height and a plastic tray at the bottom. From each soil-biochar mixture 12 replicates were prepared allowing three different experiments (untreated, "G"-treated and "N,G"-treated experiments; see below) with three replicates each at two different points in time (cf. Table 5). For the Rh soil each sample contained 100 g moist soil whereas the Oe samples only contained of 90 g moist soil because of its lower density. Each sample was then adjusted to 60 % of its max. WHC. The incubation experiments were conducted one week after sample preparation in November 2010 and in February 2011 (cf. Table 5). To examine the effect of ageing, samples measured in 2011 were stored in the dark at 24 °C. The water content was gravimetrically adjusted irregularly (every 7–20 days).

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For *incubation experiments* the steel cylinders were put in airtight glass bottles (187 ml headspace volume) and the headspace gas concentrations were analysed 3 times in the first and last four hours of a total incubation time of 22 h. The exit of each reactor was equipped with a septum for syringe sampling. Incubation temperature was 24 °C (room temperature). The gas samples were analysed by a gas chromatograph (GC, SRI 8610C) equipped with an electron capture detector (ECD), a pre-column (1 m × 8″ Restek packed Haysep-A 80/100) to separate water vapour and the analytical column (3 m × 8″ supelco porapak 80/100) to separate CO_2 and N_2O. N_2 was used as carrier gas. Depending on the concentrations levels of N_2O and CO_2 the gas sample volume was chosen (0.5 or 0.1 ml) to reduce peak overlapping. The reduction factor (1 and 5, respectively) was multiplied with the resulting concentration, assuming a linear behaviour of the ECD signal. The ECD was calibrated using different standard gas mixtures (CO_2, N_2O, and N_2) in the expected concentration ranges.

Three different *soil treatments* were conducted. (1) no addition (untreated), (2) addition of glucose at a rate of 300 kg C ha^{-1} ("G"-treated), and (3) addition of potassium nitrate and glucose at a rate of 30 kg N ha^{-1} and 300 kg C ha^{-1}, respectively, ("N,G"-treated) in two steps (one day between the applications). The amount of N applied corresponds to the amount of N which is normally applied by Swiss farmers as mineral fertilizer to grasslands after cut. The three treatments aim to represent typical field conditions, namely background fluxes with low levels of available C and N (untreated), situations of high residue accrual ("G") and application of slurry ("N,G").

In experiment (1) soil samples were measured at 60 % of max. WHC. In experiment (2) and (3) the amount of water to dissolve the glucose and nitrate, respectively, was chosen that the sample's water content reached 80 % max. WHC. Note that the emissions revealed from the "G"- and "N,G"-treated samples are not directly comparable to the untreated experiment, since the max. WHC was set to 80 % compared to 60 % in the untreated experiment.

Fluxes of each sample were calculated by applying linear regression trough three measurement points at the beginning and trough three measurement points after 22 h

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(results in ppm h^{-1} for CO_2 and N_2O fluxes). The linearity of soil gas emission was tested in separated experiments, where the samples were continuously measured for 22 h (data not shown). The flux (g m^{-2} h^{-1}) was calculated using the formula:

$$\text{flux} = \frac{\text{headspace volume} \times \text{slope} \times \text{mole weight}}{\text{sample area} \times \text{molar volume}\,(T = 25\,^{\circ}\text{C},\ p = 1\,\text{atm})} \quad (1)$$

NO_3^- and NH_4^+ concentrations were measured to estimate the availability of N in the samples. Measurements were done in February 2011 from stored (4 °C) material representing fresh soil-biochar samples (named as 2010; cf. Table 5) and from untreated samples of the 2011 experiment (stored at 24 °C since November 2010) representing aged samples (named as 2011; cf. Table 5). Concentrations were measured after dissolving 20 g moist soil in 80 ml 0.01 M $CaCl_2$-solution. Samples were shaken for 30 min and afterwards filtered (Low Nitrogen P, K free Filter Paper; Cod: PL 1290150; Filtros Anoia, S.A. Barcelona). The filtrate was analysed by HACH Lange spectrometer equipment (LCK 304 with Ammonium cuvette test measuring range 0.015–2 mg l^{-1} NH_4^+-N and LCK 339 with Nitrate cuvette test measuring range 0.23–13.5 mg l^{-1} NO_3^--N) in a DR 2800 spectrometer.

For *statistical analysis* of differences in N_2O and CO_2 emissions, respectively, the treatments and gas species were analysed separately by ANOVA using three factors soil (Oe, Rh), biochar (control, green waste, coffee grounds), and time (2010, 2011) and their interactions. N_2O and CO_2 fluxes were not normally distributed. Therefore, flux data sets were transformed by Yeo-Johnson power transformation (Yeo and Johnson, 2000) and tested for normality by six different tests (Lilliefors Kolmogorov-Smirnov test, Shapiro-Francia test, Shapiro-Wilk test, Anderson-Darling test, Cramer-von Mises test, and Pearson chi-square test). The transformed data was assumed to be normally distributed if at least three of the tests showed reasonable P values (> 0.5) and the ANOVA was performed in R. The CO_2 data sets from untreated experiment was square root transformed to fulfil the above described test for normality. The CO_2 data sets from "N,G"-experiment was square root transformed but did not fulfil the above test

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for normality. However, this was the best possible transformation found and hence the measurements were included in the analysis. The statistical analysis for NO_3^- and NH_4^+ concentration was performed with untransformed data using the Welch Two Sample two sided t-test ($\alpha = 0.05$) in R.

3 Results

Figure 1 shows the N_2O (left) and CO_2 (right) fluxes from both soils (Oe, Rh) and biochar combinations (control (c), gw, cg) depending on point in time (2010 left, 2011 right) and the different treatments (rows). Table 6 lists the reduction in percentage of N_2O and CO_2 fluxes relative to control samples. The data which underlies the Fig. 1 are listed in Tables 9–14 at the end of the paper.

Soil incubation experiments with *untreated* char-soil mixtures at 60 % of max. WHC emitted little N_2O and CO_2 (Fig. 1a,b) with large variations. The emissions from the Oe samples were in the range of static chamber measurements at the Oensingen site (Flechard et al., 2005).

Both N_2O and CO_2 emissions in the untreated samples measured in November 2010 ("2010 samples") were higher than from the aged samples. N_2O fluxes were not significantly altered by biochar amendment (Table 7) but biochar reduced CO_2 emission in the "2010 samples". Relative reductions in N_2O fluxes ranged from 7 to 48 % for Oe soil. However, with biochar the N_2O flux even increased in one experiment (Rh-gw, 2011). The other experiments showed relative reductions in the range of 81 to 432 %. With cg-biochar the relative emission reduction increased with ageing for both soils but not consistently with gw-biochar (Table 6). CO_2 emissions were also low, indicating a low micro-organism activity. The aged samples showed lower emissions compared to the "2010 samples". All CO_2 flux reductions were smaller after the ageing of three months.

Emissions from *glucose treated* samples (at 80 % max. WHC) were generally much higher than from untreated samples (Fig. 1c,d). Without biochar Rh soil emitted less N_2O than Oe soil. N_2O emissions from aged samples were significantly higher

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compared to 2010 (Oe soil), but were similar for Rh soils at both points in time. Also CO_2 emissions were in the same order of magnitude at both points in time.

Relative to the control, biochar application systematically and significantly reduced N_2O from both soils by 40 to 98 % (Table 6). No clear reduction pattern was found for CO_2. In both "G"-treated soils but more pronounced for Oe soil relative N_2O reductions decreased from 2010 to 2011. However, absolute N_2O emissions were much higher in 2011. The relative reduction in CO_2 emissions were lower than for N_2O and in two cases higher fluxes were measured from biochar amended compared to the control soil (cf. Table 6: Oe-gw and Oe-cg, 2011).

Samples treated with *glucose and potassium nitrate* emitted significantly less N_2O in biochar amended samples compared to controls. Decreases in CO_2 emissions were less pronounced (Fig. 1e,f). The relative reductions ranged from 29 to 79 % for N_2O fluxes whereas they ranged from −3 to 44 % for CO_2. For Oe soils the relative reduction of N_2O fluxes increased with ageing, whereas for Rh soil the ageing effect was biochar dependent.

Table 7 shows a summary of the statistical analysis of N_2O and CO_2 fluxes (ANOVAs). No significant difference in N_2O fluxes due to biochar incorporation from the untreated samples was found. The "G"- and "N,G"-treated experiments showed both significant differences in N_2O emissions due to biochar application. For CO_2, although the ANOVA showed significant differences, there was not always a reduction in the emissions.

At the beginning of the experiment NO_3^- *concentrations* from untreated samples were higher in Oe soil than in Rh soil whereas NH_4^+ *concentrations* did not differ between soils (Table 8). All samples showed a positive net N mineralization over time. Whereas the accumulation was significant for NO_3^-, the accumulation of NH_4^+ was less pronounced. The NO_3^- mineralization rate was not consistent for the control and charred samples. The highest NO_3^- mineralization rate was found for Oe soil with cg-biochar (factor 12), but the absolute amount was still below that of the control sample after ageing. Overall the nitrogen availability has increased in the absence of plants.

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Compared to the control the initial NO_3^- concentrations in the soil-biochar mixtures were significantly lowered due to biochar addition whereas NH_4^+ concentrations were only significantly lowered in the Rh-cg sample. The NH_4^+ concentrations were similar in all soil-biochar mixtures.

4 Discussions

Biochar reduced N_2O emissions from two intensively managed temperate agricultural soils over a wide range of emission rates. For stimulation treatments ("G" and "N,G") relative reduction were highest when emissions from the control (i.e. without biochar) soils were small (62–98 % at control flux rates of up to 5 mg N_2O m^{-2} h^{-1}) and declined to values of around 29–56 % at high emissions of over 18 mg N_2O m^{-2} h^{-1} as induced by "N,G"-treatment in Oe soil and by "G"-treatment in Oe soil in 2011. Emission reductions were also observed for untreated samples albeit many of the emissions in this treatment were not significantly different from zero. CO_2 emissions were slightly reduced, too, but not in all cases.

N_2O emissions were systematically higher in the Oe soil which we mainly attribute to its higher clay content and its higher proportion of water-saturated micropores then the loamy Rh soil. In addition, the Oe soil had a higher potential for net N mineralization (Table 8) due to its higher organic matter content. The overall higher emission of the Oe soil enabled a 2.5 to 7.0 fold stronger absolute emission reduction with biochar ("G"- and "N,G"-treatments). Hence the soil type itself played an important role for the reduction capacity.

Across all treatments neither the absolute nor the relative reduction effect by biochar application declined significantly during the experiment. But the "ageing" of gw-biochar lowered the reduction capacity in both soils for the untreated and "G"-treated experiment but not in the "N,G"-treated experiment. The ageing of the cg-biochar showed no consistent pattern.

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In our experiment the biochar application rate was smaller (2 % w/w equal to about 8 t ha^{-1}) compared to other studies with 10 % w/w (Spokas and Reicosky, 2009), 100 and 200 t ha^{-1} (Kammann et al., 2011) or 2–60 % w/w (Spokas et al., 2009). This suggests that small amounts of biochar at least initially reduce N_2O emissions, albeit it is not known whether the char has to be re-applied if the effect should last for longer time periods.

4.1 Effects of N and C availability on GHG emissions

The very low N_2O emissions measured from untreated samples indicated that denitrification was strongly C-limited. The accumulation of NO_3^- in the 2011 samples did not lead to higher N_2O emissions either and therefore supports the hypothesis of C-limitation being the cause for the small emissions. The results also showed that nitrification was active, whereas denitrification was most likely on a very low level during the ageing process. The low N_2O emissions from untreated samples may be related to a low abundance of denitrifiers but an increased population can be assumed as soon as the C-limitation was reduced by adding glucose, indicated by the significantly increased absolute N_2O and CO_2 emissions (one order of magnitude higher than in the untreated samples). When the C-limitation was overcome the N_2O emissions correlated with NO_3^- concentration; lower NO_3^- concentrations in 2010 corresponded to lower absolute N_2O emissions compared to 2011 when both, concentrations and emissions, were higher. Hence biochars had the ability to reduce available N in soil and/or the use of C as energy source for micro-organisms. The results further suggest that gw-biochar was more effective in reducing N_2O emissions at low NO_3^--concentrations whereas cg-biochar reduced N_2O emissions more effectively after NO_3^- addition and accumulation within three months. We speculate that the main reason for the observed reduction in N_2O and CO_2 emissions after biochar application was the reduced microbial availability of N and C.

Whereas in "G"-treatments cg-biochar always reduced N_2O emissions more than gw-biochar, the latter was more efficient in the "N,C"-treatment ("2011 samples"). Hence

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the reduction depended both on the type of biochar and the superimposed treatment. The single high reduction of 432 % occurred relative to a control flux of around zero, resulting in a very high relative reduction efficiency and is therefore of very limited relevance for the effect of biochar amendment on N_2O emissions.

Biochar limited the availability of N in soil by the adsorption of NO_3^- and NH_4^+ at its high surface area (Mizuta et al., 2004; Singh et al., 2010) and hence decrease N_2O emissions due to a smaller amount of N available for denitrification and nitrification. The NO_3^- concentration measured by $CaCl_2$-extraction does not necessarily represents the available NO_3^- for denitrification. However, the effect of biochar on N availability can best be studied in the experiments without additional treatment. In 2010 the biochar amendment significantly decreased NO_3^- availability but the decrease did not correspond to the surface area of the char. The specific surface area was measured by classical N_2 adsorption and is therefore a weak predictor for the capacity to adsorb ions (in this study NO_3^- and NH_4^+). Furthermore the specific surface area of the mixed samples was calculated but not measured. It is possible that the incorporation of biochar into soils has not an additive effect on the specific surface area. Therefore any conclusions regarding the adsorption effect drawn from this experiments are necessarily speculative. The data showed that adsorption reduced available NO_3^- directly after biochar incorporation, but it could not explain the still high emission reductions when high NO_3^- concentrations were present. Mizuta et al. (2004) measured a maximal adsorption capacity of bamboo powder charcoal of about 1 mg NO_3^- g^{-1} biochar at 15–20 °C. According to this number the supplementation of 2 % w/w biochar should potentially decrease the amount of NO_3^- by about 20 mg kg^{-1} soil. This is within the range of our NO_3^- measurements (Table 8).

With ageing the effect of adsorbing NO_3^- at the biochar's surface seems to diminish as only the cg-samples had significantly lower NO_3^- concentrations.

Biochar could also decrease C-substrate availability through adsorption and stabilisation of labile C at its surface (Clough and Condron, 2010). The experiment did not show a consistent reduction of CO_2 in the biochar amended soils. The experiments

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without glucose addition showed that C was the primary limiting factor for soil respiration and N_2O production. Kimetu and Lehmann (2010) suggested biochar to enhance the incorporation of labile C to the intra-aggregate fraction, hence C is stabilised more effectively. Our results contradicts this hypothesis since there was no consistent reduction in CO_2 fluxes. Also the presumably limited adsorption capacity of biochar for dissolved carbon, which is continuously formed during decomposition, may not be sufficient to sustain the reduction effect for several months.

However, there were interactions between biochar type and N and C availability indicating that the type of biochar is an important control depending on the environmental conditions.

4.2 Effect of pH change

Alkaline biochars increased pH of the soil by 2–5 tenth pH units. Increased pH promotes complete reduction to N_2 (Stevens et al., 1998) by increasing the activity of N_2O-reductase enzymes (Cavigelli and Robertson, 2000). Other steps of nitrification and denitrification could possibly be influenced by pH changes, too. Adsorption behaviour as well depends on pH. Van Zwieten et al. (2010) and Klemedtsson et al. (2010) support the hypothesis of pH being responsible for N_2O reductions. In our experiments the change in pH after biochar application was significantly positively correlated to the relative reduction in N_2O emissions ($r = 0.56$, $p < 0.05$; "G" and "G,N" only). Because the biochar effect was not tested against a pH-increase without biochar no conclusion can be drawn whether reduced N_2O fluxes were due to a biochar induced pH changes but the results do not falsify the pH hypothesis.

4.3 Comparison to field data

The three different treatments (untreated, "G"- and "N,G"-treated) can be related to season specific conditions on the agricultural field. Experiments without C or N addition showed a very low soil microbial activity. Such conditions refer to cold seasons

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and depleted labile C and N with little decomposition and small N_2O emissions. The fluxes measured form untreated samples in the lab were low as compared to stimulation treatments but compared well to baseline emissions measured by static chamber flux measurements at the same site in Oensingen (Flechard et al., 2005). Also fluxes in the "N,G"-treatment which may reflect situation when mineral fertilizer or cattle slurry is applied compared favourable to emission peaks in the Oensingen field. Flechard et al. (2005) measured monthly maxima of up to 8.6 mg N_2O m^{-2} h^{-1} which is similar to the maximum emissions of around 20 mg N_2O m^{-2} h^{-1}. These authors also showed that major event of N_2O emissions counting for cumulative losses in Oensingen occur after application of mineral fertiliser, cattle slurry or following grass cutting or freeze/thaw processes. With the addition of glucose a situation with higher microbial activity was simulated, comparable to field situations when larger quantities of residues become available such as after a cut. The soil samples in the laboratory contained little plant material hence the available substrate for micro-organisms was very small. The addition of glucose may correspond to a more typical and higher availability of crop/plant residues (Gillam et al., 2008) and also higher temperatures. According to Gillam et al. (2008) C from glucose is up to 50 % more available for micro-organisms as from clover residues, hence higher emissions after glucose addition compared to field emissions are expected. Under field conditions emissions are more flattened because most of the labile soil organic matter or plant residues first needs to be depolymerised. N_2O peaks after cutting could have similar impacts.

According to our data, emission peaks after cutting or slurry application may be reduced by around 40–60 % if the magnitude of the biochar effect in the lab is similar in the field.

When converting the laboratory results to the field situations strongest relative reductions are likely in situations when N is limited (baseline emissions) but also emission peaks may be reduced substantially. However, because of the strong competition of plants for available N and their effect on abiotic and chemical soil conditions, extrapolation to the real world must be done with caution and future field studies are strongly

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needed. Although there is an increasing number of field studies until now the focus did not lie in temperate regions (e.g, Scheer et al., 2011). This is required given the different mineralogy and fertility of temperate vs. tropical soils. Furthermore, the effect of ageing in the lab at constant 24 °C does not represent field conditions and the reduction effect in the field may be less or stronger after the ageing of the biochar.

5 Conclusions

The present lab study showed that biochar has the potential to substantially reduce N_2O emissions from temperate soil without a diminishment of the effect over three months. Because the reduction depends on soil type, biochar type, and substrate availability for micro-organisms a classification of different soil and biochar types regarding their reduction potential is needed. Additionally, although biochar showed a high potential to considerably reduce amounts of N_2O in the lab, the observed GHG reductions cannot be transferred to field conditions easily. Mainly the ageing effect at a constant temperature of 24 °C does not represent field conditions. Furthermore the experiment was conducted without vegetation, which strongly controls the availability of NO_3^- and NH_4^+ during the growing season. Therefore field studies comparing N_2O fluxes from plots where no biochar is applied to plots with incorporated biochar are suggested as a next step to get more details whether biochar can be seen as mitigation option for N_2O emission in temperate agriculture.

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Table 1. Biochar properties: specific surface in $m^2\,g^{-1}$ dry matter, carbon and nitrogen content in percent of dry matter, C/N-ratio (by mass), pH, and temperature during pyrolysis.

	surface $m^2\,g^{-1}$	C %	N %	C/N	pH	T_{max} °C
green waste (gw)	177.1	54.9	0.7	79.7	9.1	750
coffee grounds (cg)	24.2	66.5	2.6	25.5	9.4	900–950

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Table 2. Elemental composition of green waste (gw) and coffee grounds (cg) biochar per dry matter as measured by X-ray fluorescence.

	P $g\,kg^{-1}$	K $g\,kg^{-1}$	Ca $g\,kg^{-1}$	Mg $g\,kg^{-1}$	Na $mg\,kg^{-1}$	Cu $mg\,kg^{-1}$	
gw	2.44	9.34	68.5	5.37	736	40.3	
cg	4.92	9.46	25.2	7.83	982	56.8	
	Zn $mg\,kg^{-1}$	B $mg\,kg^{-1}$	Mn $mg\,kg^{-1}$	Mo $\mu g\,kg^{-1}$	S $mg\,kg^{-1}$	Fe $g\,kg^{-1}$	Ni $mg\,kg^{-1}$
gw	99.9	47.9	398	1424	1100	9.27	23.1
cg	142.4	24.6	424	593	1092	3.27	26.2

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Table 3. Soil properties of the Oensingen and Reckenholz soils (C/N ratio by mass).

	spec. surface $m^2\ g^{-1}$	clay content w%	SOC %	C/N	pH
Oensingen (Oe)	14.85	44	3.0	9.5	7.0
Reckenholz (Rh)	5.81	19	1.5	8	6.3

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Table 4. Sample's label, specific surface area, maximum water holding capacity and pH for all sample combinations.

label	soil	biochar	spec. surface $m^2\ g^{-1}$	max. WHC % dry matter	pH
Oe-c	Oensingen	none = control	14.85	94.1	7.0
Oe-gw	Oensingen	green waste	18.03*	98.3	7.2
Oe-cf	Oensingen	coffee grounds	15.11*	95.3	7.3
Rh-c	Reckenholz	none = control	5.81	55.8	6.3
Rh-gw	Reckenholz	green waste	8.14*	59.1	6.8
Rh-cg	Reckenholz	coffee grounds	6.17*	58.4	6.7

* specific surfaces of mixed samples were calculated by: $\frac{A_{\text{biochar}} \times m_{\text{biochar}} + A_{\text{soil}} \times m_{\text{soil}}}{m_{\text{biochar}} + m_{\text{soil}}}$.

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Table 5. Overview of sample storage and measurements.

date	sample group A	sample group B	rest material
Oct 2010	soil collections, mixing with biochars		
	stored at 24 °C	stored at 24 °C	stored at 4 °C
Nov 2010	**experiments 2010**	|	|
	untreated	|	|
	"G"-treated	|	|
	"N,G"-treated	|	|
Feb 2011		**experiments 2011**	|
		untreated	|
		"G"-treated	|
		"N,G"-treated	|
Feb 2011		**NO_3^-/NH_4^+ meas.**	**NO_3^-/NH_4^+ meas.**
		i.e. conc. in 2011	i.e. conc. in 2010

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Table 6. Absolute and relative reductions of CO_2 and N_2O fluxes from two biochar amended soils. Relative reductions are given as percentage of N_2O and CO_2 flux relative to control soils, respectively. A value less than zero means an increased flux compared to the control.

			Reduction of N_2O fluxes				Reduction of CO_2 fluxes			
			green waste		coffee grounds		green waste		coffee grounds	
soil	treat.	year	$\mu g\,m^{-2}\,h^{-1}$	%	$\mu g\,m^{-2}\,h^{-1}$	%	$mg\,m^{-2}\,h^{-1}$	%	$mg\,m^{-2}\,h^{-1}$	%
Oe	–	2010	0.4	48	0.1	7	11.7	35	12.3	37
Oe	–	2011	0.1	35	0.04	22	0.3	3	−0.1	−0.9
Oe	G	2010	3111	67	4425	96	4.2	1	160.5	26
Oe	G	2011	7423	40	10 332	56	−3.1	−0.6	−15.6	−3
Oe	N,G	2010	10 284	46	6493	29	467.2	44	73.4	7
Oe	N,G	2011	11 858	54	9095	41	−3.5	−0.6	−21.2	−3
Rh	–	2010	0.4	81	0.4	101	14.2	46	9.1	30
Rh	–	2011	−0.6	−8119	0.03	432	5.1	36	4.1	29
Rh	G	2010	1109	91	1199	98	80.6	14	235.1	41
Rh	G	2011	1305	82	1474	93	214.8	29	156.7	21
Rh	N,G	2010	2209	62	2554	72	150.9	22	119.6	17
Rh	N,G	2011	3170	79	2519	63	209.9	30	70.3	10

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Table 7. Summary of the ANOVA output for all measurements. The detailed ANOVAs are found in Tables 15–20 at the end of the paper.

	N_2O fluxes			CO_2 fluxes		
factors/treat.	untreat.	"G"-treat.	"N,G"-treat.	untreat.	"G"-treat.	"N,G"-treat.
soil		**	**			**
char		**	**	**	**	**
year		**		**	**	**
soil : char		**	**		**	**
soil : year	*	**	*		**	**
char : year		**	**		**	**
soil : char : year			**		*	**

signif. levels:
"**" < 0.01
"*" < 0.05
" " < 1

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Table 8. Nitrate (NO_3^-) and ammonia (NH_4^+) contents in mg per kg dry matter in the untreated soil samples.

sample	NO_3^- 2010		NO_3^- 2011		NH_4^+ 2010		NH_4^+ 2011	
	mean	sd	mean	sd	mean	sd	mean	sd
Oe-c	94.0	0.6	693.2 [b]	52.8	0.8	0.3	3.1	2.6
Oe-gw	71.1[a]	4.4	620.4 [b]	4.2	0.7	0.2	2.4	0.8
Oe-cg	42.9[a]	1.0	530.2[a,b]	14.2	0.7	0.2	1.5	0.5
Rh-c	80.6	18.3	263.7 [b]	2.2	1.1	0.1	1.9	0.6
Rh-gw	53.2	2.9	318.8 [b]	27.8	0.9	0.1	1.9	0.7
Rh-cg	22.7[a]	0.5	194.2[a,b]	4.6	0.5[a]	0.1	1.9	0.9

[a] indicates a significant concentration difference of soil-biochar mixtures compared to control sample within the same year.
[b] indicates a significant accumulation after the ageing of a sample.

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Table 9. N_2O and CO_2 fluxes from untreated soil samples 2010.

factors				CO_2	N_2O
year	soil	biochar	treat.	mg m^{-2} h^{-1}	µg m^{-2} h^{-1}
2010	Oe	control	-	25.26	0.44
2010	Oe	control	-	24.26	1.20
2010	Oe	control	-	19.73	−0.01
2010	Oe	control	-	45.58	1.34
2010	Oe	control	-	43.06	1.24
2010	Oe	control	-	40.94	1.02
2010	Rh	control	-	27.89	1.12
2010	Rh	control	-	23.61	−0.72
2010	Rh	control	-	23.60	−0.52
2010	Rh	control	-	37.79	1.96
2010	Rh	control	-	36.23	0.45
2010	Rh	control	-	34.86	0.37
2010	Oe	green waste	-	10.22	−0.41
2010	Oe	green waste	-	14.06	0.02
2010	Oe	green waste	-	14.34	0.17
2010	Oe	green waste	-	29.81	0.9
2010	Oe	green waste	-	28.99	0.83
2010	Oe	green waste	-	31.10	1.22
2010	Rh	green waste	-	11.54	−0.53
2010	Rh	green waste	-	11.50	−0.17
2010	Rh	green waste	-	11.43	−0.36
2010	Rh	green waste	-	24.68	0.64
2010	Rh	green waste	-	19.13	0.46
2010	Rh	green waste	-	20.75	0.47
2010	Oe	coffee grounds	-	9.98	−0.08
2010	Oe	coffee grounds	-	11.15	−0.08
2010	Oe	coffee grounds	-	10.49	−0.08
2010	Oe	coffee grounds	-	31.21	2.60
2010	Oe	coffee grounds	-	31.85	1.72
2010	Oe	coffee grounds	-	30.31	0.76
2010	Rh	coffee grounds	-	16.56	−0.50
2010	Rh	coffee grounds	-	13.35	−0.52
2010	Rh	coffee grounds	-	12.64	−0.72
2010	Rh	coffee grounds	-	31.43	0.53
2010	Rh	coffee grounds	-	28.30	0.62
2010	Rh	coffee grounds	-	27.13	0.57

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Table 10. N_2O and CO_2 fluxes from "G"-treated soil samples 2010.

factors year	soil	biochar	treat.	CO_2 mg m^{-2} h^{-1}	N_2O µg m^{-2} h^{-1}
2010	Oe	control	G	616.45	4966.97
2010	Oe	control	G	608.64	5047.04
2010	Oe	control	G	607.81	3822.26
2010	Rh	control	G	586.43	1433.87
2010	Rh	control	G	574.00	1013.44
2010	Rh	control	G	Na	Na
2010	Oe	green waste	G	588.46	653.06
2010	Oe	green waste	G	616.97	1687.63
2010	Oe	green waste	G	614.73	2162.7
2010	Rh	green waste	G	528.34	107.71
2010	Rh	green waste	G	512.95	129.89
2010	Rh	green waste	G	457.55	105.00
2010	Oe	coffee grounds	G	439.30	186.32
2010	Oe	coffee grounds	G	463.94	199.82
2010	Oe	coffee grounds	G	448.16	173.70
2010	Rh	coffee grounds	G	330.59	21.27
2010	Rh	coffee grounds	G	351.44	36.63
2010	Rh	coffee grounds	G	353.46	16.59

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Table 11. N_2O and CO_2 fluxes from "N,G"-treated soil samples 2010.

factors year	soil	biochar	treat.	CO_2 mg m^{-2} h^{-1}	N_2O µg m^{-2} h^{-1}
2010	Oe	control	N,G	1105.63	22 666.97
2010	Oe	control	N,G	1043.53	21 949.89
2010	Oe	control	N,G	1033.06	21 783.98
2010	Rh	control	N,G	695.94	3540.02
2010	Rh	control	N,G	672.58	3567.23
2010	Rh	control	N,G	721.89	3574.83
2010	Oe	green waste	N,G	575.33	10 951.51
2010	Oe	green waste	N,G	618.16	13 168.99
2010	Oe	green waste	N,G	587.13	11 428.08
2010	Rh	green waste	N,G	553.03	1198.04
2010	Rh	green waste	N,G	527.76	1468.34
2010	Rh	green waste	N,G	557.04	1388.76
2010	Oe	coffee grounds	N,G	950.06	14 104.74
2010	Oe	coffee grounds	N,G	963.24	17 633.21
2010	Oe	coffee grounds	N,G	1048.63	15 184.03
2010	Rh	coffee grounds	N,G	590.65	893.79
2010	Rh	coffee grounds	N,G	582.08	1020.83
2010	Rh	coffee grounds	N,G	558.81	1105.14

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Table 12. N_2O and CO_2 fluxes from untreated soil samples 2011.

factors				CO_2	N_2O
year	soil	biochar	treat.	mg m^{-2} h^{-1}	µg m^{-2} h^{-1}
2011	Oe	control	-	9.32	0.36
2011	Oe	control	-	7.79	0.38
2011	Oe	control	-	7.84	0.29
2011	Oe	control	-	9.15	0.06
2011	Oe	control	-	9.22	−0.08
2011	Oe	control	-	8.84	−0.03
2011	Rh	control	-	11.73	0.12
2011	Rh	control	-	11.67	0.06
2011	Rh	control	-	15.15	0.25
2011	Rh	control	-	15.95	−0.02
2011	Rh	control	-	13.70	−0.36
2011	Rh	control	-	15.04	−0.09
2011	Oe	green waste	-	9.49	0.31
2011	Oe	green waste	-	8.88	0.19
2011	Oe	green waste	-	7.23	0.33
2011	Oe	green waste	-	9.32	−0.06
2011	Oe	green waste	-	6.77	−0.30
2011	Oe	green waste	-	8.80	0.18
2011	Rh	green waste	-	5.76	0.40
2011	Rh	green waste	-	5.88	0.16
2011	Rh	green waste	-	9.56	0.95
2011	Rh	green waste	-	10.71	0.68
2011	Rh	green waste	-	11.01	0.56
2011	Rh	green waste	-	9.95	0.83
2011	Oe	coffee grounds	-	8.2	0.28
2011	Oe	coffee grounds	-	7.89	0.31
2011	Oe	coffee grounds	-	6.99	0.06
2011	Oe	coffee grounds	-	9.68	0.08
2011	Oe	coffee grounds	-	9.97	0.01
2011	Oe	coffee grounds	-	9.9	0.03
2011	Rh	coffee grounds	-	8.76	0.15
2011	Rh	coffee grounds	-	7.33	−0.05
2011	Rh	coffee grounds	-	7.79	−0.24
2011	Rh	coffee grounds	-	9.87	−0.32
2011	Rh	coffee grounds	-	12.48	0.09
2011	Rh	coffee grounds	-	12.65	0.13

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Table 13. N_2O and CO_2 fluxes from "G"-treated samples 2011.

factors year	soil	biochar	treat.	CO_2 mg m^{-2} h^{-1}	N_2O µg m^{-2} h^{-1}
2011	Oe	control	G	464.27	17 731.64
2011	Oe	control	G	534.08	18 501.87
2011	Oe	control	G	531.85	19 250.62
2011	Rh	control	G	762.80	2021.52
2011	Rh	control	G	744.20	1347.89
2011	Rh	control	G	730.09	1388.01
2011	Oe	green waste	G	547.10	10 406.18
2011	Oe	green waste	G	475.25	12 914.79
2011	Oe	green waste	G	517.08	9894.25
2011	Rh	green waste	G	511.32	183.79
2011	Rh	green waste	G	565.12	334.73
2011	Rh	green waste	G	516.27	325.11
2011	Oe	coffee grounds	G	479.56	7721.95
2011	Oe	coffee grounds	G	530.41	5375.67
2011	Oe	coffee grounds	G	567.12	11 390.73
2011	Rh	coffee grounds	G	555.41	90.23
2011	Rh	coffee grounds	G	570.01	88.13
2011	Rh	coffee grounds	G	641.61	156.72

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Table 14. N_2O and CO_2 fluxes from "N,G"-treated samples 2011.

factors year	soil	biochar	treat.	CO_2 mg m^{-2} h^{-1}	N_2O µg m^{-2} h^{-1}
2011	Oe	control	N,G	513.51	21 497.73
2011	Oe	control	N,G	564.36	23 115.22
2011	Oe	control	N,G	565.18	21 760.07
2011	Rh	control	N,G	665.86	4462.44
2011	Rh	control	N,G	727.40	4036.54
2011	Rh	control	N,G	716.61	3549.28
2011	Oe	green waste	N,G	540.93	11 522.1
2011	Oe	green waste	N,G	538.38	9546.40
2011	Oe	green waste	N,G	574.28	9730.35
2011	Rh	green waste	N,G	514.67	1045.43
2011	Rh	green waste	N,G	481.17	794.72
2011	Rh	green waste	N,G	484.38	697.70
2011	Oe	coffee grounds	N,G	594.82	12 966.28
2011	Oe	coffee grounds	N,G	530.02	13 776.21
2011	Oe	coffee grounds	N,G	581.68	12 344.76
2011	Rh	coffee grounds	N,G	585.69	1649.46
2011	Rh	coffee grounds	N,G	665.38	1229.43
2011	Rh	coffee grounds	N,G	648.05	1611.16

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Table 15. ANOVA results for N_2O fluxes of untreated soil samples.

	Df	Sum Sq	Mean Sq	F value	Sign. level
soil	1	0.3572	0.35722	3.6915	.
char	2	0.1005	0.05027	0.5195	
year	1	0.2719	0.27189	2.8097	.
soil : char	2	0.2896	0.14479	1.4962	
soil : year	1	0.4667	0.46667	4.8226	*
char : year	2	0.3673	0.18366	1.898	
soil : char : year	2	0.0753	0.03767	0.3892	
Residuals	60	5.806	0.09677		

Signif. codes:
"*" < 0.05
"." < 0.1
" " < 1

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Table 16. ANOVA results for CO_2 fluxes of untreated soil samples.

	Df	Sum Sq	Mean Sq	F value	Sign. level
soil	1	0.101	0.101	0.205	
char	2	10.243	5.122	10.4046	***
year	1	51.4	51.4	104.4209	***
soil : char	2	0.913	0.456	0.927	
soil : year	1	1.141	1.141	2.3187	
char : year	2	3.018	1.509	3.0659	.
soil : char : year	2	0.673	0.336	0.6831	
Residuals	60	29.534	0.492		

Signif. codes:
"***" < 0.001
"." < 0.1
" " < 1

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Table 17. ANOVA results for N_2O fluxes of "G"-treated soil samples.

	Df	Sum Sq	Mean Sq	F value	Sign. level
soil	1	118.567	118.567	770.6542	***
char	2	65.272	32.636	212.1261	***
year	1	44.337	44.337	288.1819	***
soil : char	2	2.656	1.328	8.6323	**
soil : year	1	10.389	10.389	67.525	***
char : year	2	7.066	3.533	22.9645	***
soil : char : year	2	1.031	0.515	3.3502	.
Residuals	23	3.539	0.154		

Signif. codes:
"***" < 0.001
"**" < 0.01
"." < 0.1

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Table 18. ANOVA results for CO_2 fluxes of "G"-treated soil samples.

	Df	Sum Sq	Mean Sq	F value	Sign. level
soil	1	228	228	0.8623	
char	2	32 699	16 349.4	61.845	***
year	1	7412	7412.1	28.038	***
soil : char	2	12 149	6074.7	22.979	***
soil : year	1	22 383	22 383.4	84.6699	***
char : year	2	17 528	8763.8	33.151	***
soil : char : year	2	2069	1034.6	3.9134	*
Residuals	23	6080	264.4		

Signif. codes:
"***" < 0.001
"*" < 0.05
" " < 1

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Table 19. ANOVA results for N_2O fluxes of "N,G"-treated soil samples.

	Df	Sum Sq	Mean Sq	F value	Sign. level
soil	1	813.55	813.55	4332.245	***
char	2	117.29	58.64	312.2841	***
year	1	0.51	0.51	2.696	
soil : char	2	5.18	2.59	13.7812	***
soil : year	1	0.89	0.89	4.7487	*
char : year	2	3.77	1.88	10.0252	***
soil : char : year	2	3.73	1.87	9.9376	***
Residuals	24	4.51	0.19		

Signif. codes:
"***" < 0.001
"*" < 0.05
" " < 1

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Table 20. ANOVA results for CO_2 fluxes of "N,G"-treated soil samples.

	Df	Sum Sq	Mean Sq	F value	Sign. level
soil	1	32.474	32.474	94.8671	***
char	2	95.07	47.53	138.8617	***
year	1	76.82	76.82	224.4125	***
soil : char	2	6.1	3.05	8.9158	**
soil : year	1	78.49	78.49	229.2925	***
char : year	2	18.83	9.42	27.5034	***
soil : char : year	2	43.03	21.51	62.849	***
Residuals	24	8.22	0.34		

Signif. codes:
"***" < 0.001
"**" < 0.01

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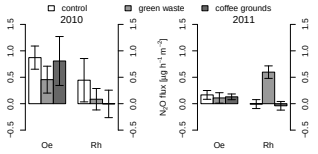
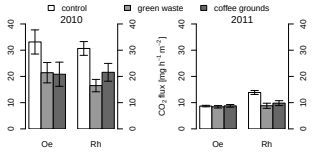
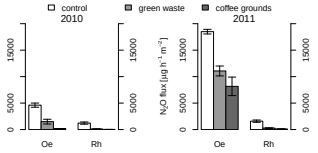


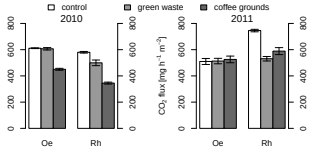
a. N_2O fluxes of untreated samples at 60 % max. WHC.



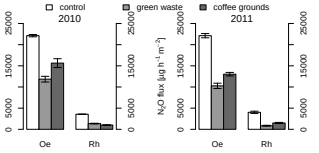
b. CO_2 fluxes of untreated samples at 60 % max. WHC.



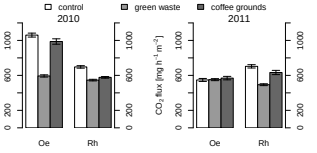
c. N_2O fluxes of "G"-treated samples at 80 % max. WHC.



d. CO_2 fluxes of "G"-treated samples at 80 % max. WHC.



e. N_2O fluxes of "N,G"-treated samples at 80 % max. WHC.



f. CO_2-fluxes of "N,G"-treated samples at 80 % max. WHC.

Fig. 1. N_2O (left column) in $\mu g\,m^{-2}\,h^{-1}$ and CO_2 (right column) fluxes in $mg\,m^{-2}\,h^{-1}$ of all sample treatments for two measurement points (2010, 2011) and two soil types (Oe, Rh). Three columns are control, green waste biochar, and coffee grounds biochar amended soils. Error bars indicate the 95 % confidence interval ($n = 6$ for untreated experiment, $n = 3$ for "G"- and "N,G"-treated experiments, respectively).

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