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Review article

BIOCHAR AS A TOOL TO REDUCE THE AGRICULTURAL GREENHOUSE-GAS BURDEN – KNOWNS, UNKNOWNS AND FUTURE RESEARCH NEEDS

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Abstract. Agriculture and land use change has significantly increased atmospheric emissions of the non-CO_2 greenhouse gases (GHG) nitrous oxide (N_2O) and methane (CH_4). Since human nutritional and bioenergy needs continue to increase, at a shrinking global land area for production, novel land management strategies are required that reduce the GHG footprint per unit of yield. Here we review the potential of biochar to reduce N_2O and CH_4 emissions from agricultural practices including potential mechanisms behind observed effects. Furthermore, we investigate alternative uses of biochar in agricultural land management that may significantly reduce the GHG-emissions-per-unit-of-product footprint, such as (i) pyrolysis of manures as hygienic alternative to direct soil application, (ii) using biochar

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as fertilizer carrier matrix for underfoot fertilization, biochar use (iii) as composting additive or (iv) as feed additive in animal husbandry or for manure treatment. We conclude that the largest future research needs lay in conducting life-cycle GHG assessments when using biochar as an on-farm management tool for nutrient-rich biomass waste streams.

Keywords: biochar, greenhouse gases (GHG), nitrous oxide (N_2O), methane (CH_4), soil aeration, nitrate, soil N transformations, GHG intensity.

Introduction: Human impact on global N_2O and CH_4 budgets and atmospheric concentrations

Human population now approaches 7.5 billion people on earth. The land area that serves human nutrition and bioenergy demands is not only limited, but declining due to soil degradation in various forms (Lal 2014; Konuma 2016; FAO 2015). The dawning perception that "fertile soils" are a finite global resource is stressed by recent land grabbing practices where wealthier countries with large populations and/or a lack of soil resources buy arable land in poorer countries. This has mostly occurred in Africa (Rulli *et al.* 2013), causing land use change to increase crop productivity, likely with consequences in terms of increasing GHG production. In fact, excessive human land use change over the past decades has contributed to the rapid, on-going increase in the atmospheric concentration of non-CO_2 greenhouse gases nitrous oxide (N_2O) and methane (CH_4) from preindustrial levels of 270–280 ppbv to 324 ppbv (N_2O) and from ~700 ppbv to 1834 ppbv (CH_4) (Myhre *et al.* 2013; Saunois *et al.* 2016).

The steep increase in atmospheric N_2O concentrations dominantly since the 1950^s is clearly the result of an increasing use of the Haber-Bosch process to generate reactive N forms from atmospheric N, plus the higher proportion of legumes on farmland compared to natural ecosystems; many important crop or fodder plants are N_2-fixing legumes (e.g. soy, pea, lentils, beans, groundnuts, clover). Global reactive-N use is now annually more than double the amount introduced by natural processes (Galloway *et al.* 2008); with increasing N fertilizer use comes the increasing likelihood of N_2O formation and atmospheric accumulation. Thus, as stated by Ravishankara *et al.* (2009), as the impact of fluorinated halocarbons decreases, N_2O will likely become the dominant O_3-depleting substance within the agricultural sector over the course of the 21st century.

Methane (CH_4) emissions have also increased, by 150% since 1750 (Myhre *et al.* 2013) to 1834 ppb in 2015 (Dlugokencky *et al.* 1994; Saunois *et al.* 2016). Human activities directly and indirectly contribute to the increased atmospheric CH_4 concentration by several pathways such as (i) expanding rice agriculture, ruminant animal husbandry and landfilling with unmanaged CH_4 emissions, (ii) thawing permafrost areas and thermocast lakes (Koven *et al.* 2011; Walter *et al.* 2006) and warming-induced changes in plant community composition e.g. expanding aerenchymal plant cover (Christensen *et al.* 2004), and (iii) "automatic" feedback effects such as rising CH_4 production under elevated, rising atmospheric CO_2 concentrations from wetlands and agricultural lands due to higher net biomass production as labile substrates for methanogenesis (Van Groenigen *et al.* 2011).

To our knowledge, the last assessment of the human impact of land-use changes and fertilizer use on global CH_4 consumption was made 20 years ago (Ojima *et al.* 1993). The authors estimated that human activities have already reduced the global net CH_4 sink capacity by 30%. Hypotheses for explaining the reduction encompass human impact on (i) soil moisture changes, (ii) reduced soil aeration via compaction / increased bulk density (both impact gas diffusivity and hence CH_4 and O_2 supply, (Castro *et al.* 1994; Hiltbrunner *et al.* 2012), and (iii) inhibition by NH_4^+ (N fertilization or reduced nitrification in acidic soils (Schnell, King 1995; Steudler *et al.* 1989). Also, (iv) shifts in the microbial community composition are hypothesized, but without conclusive evidence (Gulledge *et al.* 1997).

Over the last decades, the rising use of mineral N fertilizer (Galloway *et al.* 2008), soil degradation and forest clear-cutting, and a rising frequency of weather extremes (Hansen, Sato 2016) creating "too wet" or "too dry" soil conditions (Dijkstra *et al.* 2011) will likely further reduce the global methanotrophic CH_4 sink, and increase CH_4 and N_2O emissions. Therefore, any positive contribution that science and material use may provide agricultural soils for reducing N_2O emissions per unit yield, and in reducing either the CH_4 production and surface emissions from soils or ruminant guts (see sections 3 and 4), or by strengthening the soil CH_4 oxidation capacity (section 3.2) and its biofilter function (section 3.1), needs to be explored. Agricultural practices that utilize biochar for meeting these needs appears promising, and should be explored and developed to help lower the greenhouse gas (GHG) footprint per unit yield or bioenergy produced (also termed GHG intensity) (Wollenberg *et al.* 2016).

A reduction of N_2O and CH_4 within agroecosystems can be achieved by either increasing the per-hectare yield at unchanged GHG emission rates, by lowering the per-hectare N_2O and/or CH_4 emissions/increasing CH_4 uptake, or ideally by both. Currently literature suggests that biochar may play a role in reducing both of these GHGs; biochar can affect GHG emissions directly following its application to soils, and indirectly by adding carbonized instead of non-carbonised residue or manures which usually have higher emissions following application. Thus, the aim of this paper is to explore our current understanding

and knowledge gaps of biochar use as a tool to reduce N_2O and CH_4 emissions from agricultural land use. To this end, the biogeochemical mechanisms of formation and consumption of N_2O and CH_4 in soils and their emission to the atmosphere are presented and discussed. The topic of build-up of the soil organic carbon stocks by biochar amendment, or by reduced soil organic carbon decomposition (negative priming) is not explored here, only some rough assessments are made to illustrate potentials (e.g. for using biochar as animal feed additive).

1. Effects of biochar application to soils on N_2O emissions

1.1. Mechanisms of N_2O formation in soils and release to the atmosphere

Soils are a prominent source of N_2O emissions, especially when fertilised with organic or mineral N fertilisers. A wide range of microbial and chemical processes and pathways are responsible for these emissions (see Fig. 1), with knowledge on these pathways continuously improving. The classical view was that bacterial denitrification, the reduction of nitrate and nitrite in several steps to N_2, is the main source of N_2O from most soils, especially at intermediate water contents (Bateman, Baggs 2005). Various bacteria that are phylogenetically unrelated are capable of denitrification, with many not having the full set of enzymes for the complete pathway (Zumft 1997). This leads to the escape of intermediates, including N_2O. Under certain circumstances such as low pH and high NO_3/C_{org} ratios, the production of N_2O is favoured compared to the final production of N_2. The reduction of N_2O to N_2 is performed by the enzyme nitrous oxide reductase, which is encoded by the nosZ gene in denitrifying bacteria. In a laboratory study, lower N_2O emissions were inversely correlated to the nosZ gene expression (Harter *et al.* 2014).

Meanwhile, we know that bacterial nitrifiers may dominate N_2O emission from some soils, e.g. by the pathway nitrifier denitrification (Kool *et al.* 2011a). Bacterial nitrifiers use ammonia as a substrate and reduce intermediately produced nitrite in a comparable way as in denitrification. Nitrifiers have been found to be also able to use nitrite provided exogenously in incubation studies under aerobic conditions (Shaw *et al.* 2006). Fungi may play a dominant role for N_2O production from soils, either by codenitrification or by fungal denitrification (Laughlin, Stevens 2002; Rohe *et al.* 2014). The role of archaea for N_2O production from soils is still unknown, but there is evidence of potentially significant archaeal contributions (Jung *et al.* 2014), although the responsible pathways are yet unresolved (Jung *et al.* 2014; Stieglmeier *et al.* 2014).

As these processes may take place simultaneously in different soil microsites, it is not straightforward to distinguish among them. Several methods have recently been developed that try to unravel the sources of N_2O, including stable isotopes (Sutka *et al.* 2006; Kool *et al.* 2011b; Rohe *et al.* 2014; Lewicka-Szczebak *et al.* 2016) and molecular or modelling approaches (Rütting, Müller 2007; Kozlowski *et al.* 2014; Perez-Garcia *et al.* 2014; Snider *et al.* 2015). So far, no single method has offered a complete picture of the diverse N_2O producing pathways and a combination of methods seems most promising.

1.2. Impact of biochar on soil N_2O emissions: frequent observations and assumed mechanisms

One of the first biochar experiments reporting reduced N_2O emissions was presented in the 3rd USDA Symposium on greenhouse gases and carbon sequestration in agriculture and forestry (Rondon *et al.* 2005). A significant decrease in N_2O emissions was observed in pots planted with soybean and grass in a greenhouse experiment. However,

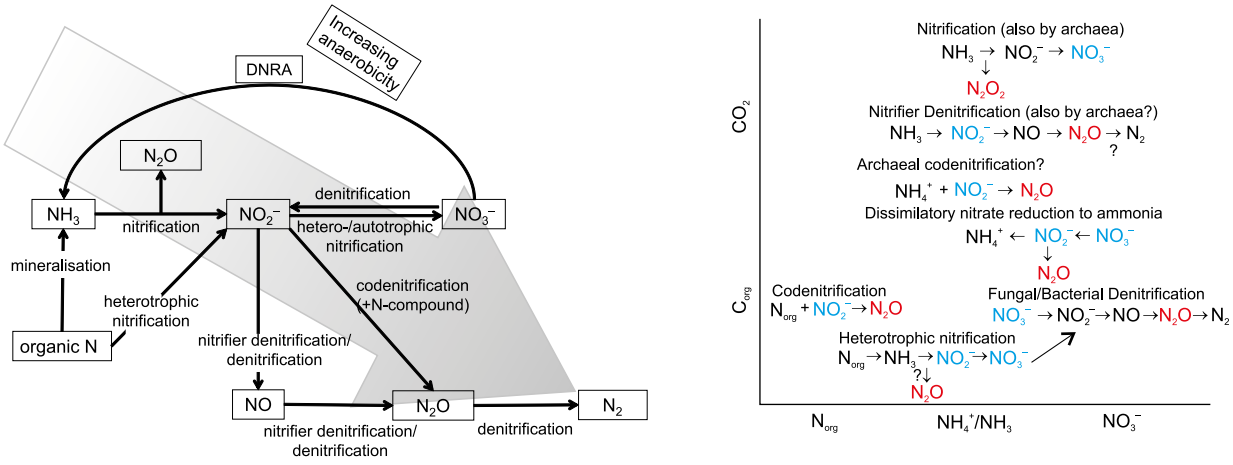

Fig. 1. a) Processes and b) pathways of N conversions associated with N_2O emissions in soils in relation to N transformations (A) and N and C substrates (B). (Figure credit: N. Wrage-Mönnig)

this finding went unnoticed for several years and only after the pioneering studies by Yanai *et al.* (2007), Spokas *et al.* (2009) and van Zwieten *et al.* (2009), the number of publications on this topic started to rise. Hence, a new field of research was established, exploring a potential win-win situation: biochar not only sequestered carbon but also had the potential to decrease non-CO_2 GHG emissions.

To date, the hypotheses for biochar's impact in char-soil mixtures on N_2O emissions has been linked to biochar properties, the soil and the environmental conditions such as temperature and precipitation (Spokas, Reicoscoky 2009; Dicke *et al.* 2015). Studies have mostly been carried out in the lab using sieved/disturbed soil samples wetted either to the same gravimetric moisture, or to the same water-filled pore space, water holding capacity or water potential. Other investigations in combination with plant growth in the greenhouse or under less controlled conditions in the field have also shown that biochar may affect the soil N_2O emissions (e.g. Taghizadeh-Toosi *et al.* 2011; Schimmelpfennig *et al.* 2014; Kammann *et al.* 2012; Deng *et al.* 2015; Hüppi *et al.* 2015). In the presence of N_2O-producing earthworms (soil fauna interactions), N_2O emissions were also reduced by biochar application (Augustenborg *et al.* 2012; Bamminger *et al.* 2014). However, in some studies no difference between biochar and control treatments was observed (Scheer *et al.* 2011; Sánchez-García *et al.* 2016) or N_2O emissions were increased from biochar amended soils (e.g. Spokas, Reicoscoky 2009; Clough *et al.* 2010; Saarnio *et al.* 2013; Troy *et al.* 2013). However, laboratory results cannot be generalised to field expectations. In field trials, often no statistical differences are observed between biochar and control treatments following field application of biochar (Castaldi *et al.* 2011; Jones *et al.* 2012; Karhu *et al.* 2011; Scheer *et al.* 2011; Schimmelpfennig *et al.* 2014; Suddick, Six 2013; Dicke *et al.* 2015). One potential reason for no significant biochar effects on N_2O emissions may be the application dose, less homogeneous particle distribution and greater soil (and plant) heterogeneity in fields resulting in high variability in N_2O fluxes (large error bars, e.g. Hüppi *et al.* 2015).

Nevertheless, overall, meta-analyses confirmed that N_2O emissions are reduced with biochar application rates of 1–2% by weight (van Zwieten *et al.* 2015; Cayuela *et al.* 2014). In spite of the extensive literature published during the past several years on the topic, knowing if a biochar will be effective in mitigating N_2O emissions in a certain agricultural field is still highly unpredictable. Thus, most research efforts are now directed towards achieving the largest N_2O emission reductions (what type of biochar to use in what soils) by analysing the mechanisms involved. Many studies have shown that biochar N_2O mitigation capacity will depend not only on the characteristics of the biochar, but also on the type of soil and predominant environmental conditions (Cayuela *et al.* 2013; Malghani *et al.* 2013; Nelissen *et al.* 2014). A remarkable finding was that, under identical environmental conditions, the same biochar could increase emissions in one soil and decrease emissions in another (Yoo, Kang 2012; Sánchez-García *et al.* 2014). This fact seems to be linked to diverse N_2O formation mechanisms operating in different soils, of which biochar might be affecting differently. In this sense, knowing how biochar interacts with the key microbial pathways regulating N_2O formation and consumption in soil is crucial for developing and implementing effective mitigation strategies. However, the number of studies looking at specific N_2O formation pathways is still very limited and the mechanisms mediating N_2O suppression are still unresolved.

To date, most N_2O-biochar studies selected certain environmental conditions and assumed or speculated the predominant N_2O formation mechanisms. For instance, studies at high water-filled pore space anticipated that the main N_2O formation pathway would be heterotrophic denitrification. However, this reasoning has frequently been shown to be incorrect. In complex soil environments, ammonia oxidation and nitrifier denitrification generally coexist with heterotrophic denitrification (Hu *et al.* 2015) and the proportion of N_2O produced in each pathway depends on many factors, not just water-filled pore space (Wrage *et al.* 2001; Butterbach-Bahl *et al.* 2013). Therefore, studies that really differentiate among N_2O produced by different sources after addition of biochars are still urgently needed.

1.2.1. What do we know about the impact of biochar on denitrification N_2O*?*

Denitrification is classically the most well-known mechanism leading to N_2O emissions and to date, also the most investigated in biochar studies. Biochar might interact with denitrification in different ways. Biochar might directly stimulate or suppress total denitrification, i.e. the amount of N that goes to gaseous form (N_2 + N_2O + NO). The impact of biochar on total denitrification has been barely studied and the results are inconclusive. For instance, using stable isotope enrichment, Cayuela *et al.* (2013) analyzed the flux of total N denitrified (N_2 + N_2O) at the peak of N_2O emissions and found that biochar decreased the total denitrificatory N efflux in 9 out of 15 soils, but significantly increased the flux in two soils. Obia *et al.* (2015) measured NO, N_2O and N_2 by high resolution gas kinetics under strictly anaerobic conditions and calculated the maximum induced denitrification rate, which was found to increase with one type of biochar (cacao shell), but not with another biochar (rice husk) in an acidic soil. By using the acetylene inhibition technique, Ameloot *et al.* (2016) found a general decrease in total denitrified N (N_2 + N_2O) with biochar in a neutral soil. Biochar might therefore

decrease or increase total denitrification depending on the type of soil. More studies are needed to understand the mechanisms behind these observations.

A decrease in total denitrification was initially attributed to improved soil aeration following biochar addition (Zhang *et al.* 2010), a hypothesis that has been rebutted by Case *et al.* (2012) who demonstrated that soil aeration played a minimal role in N_2O mitigation. Furthermore, several studies used adjusted water contents to account for increased water holding capacities that often arise when biochar is mixed into the soil (e.g. light-weight porous biochars in sandy soils), to render simple aeration effects unlikely (e.g. Kammann *et al.* 2012); still these studies observed significant N_2O emission reductions. Another hypothesis suggested a general decrease in soil microbial activity as a consequence of toxic compounds present in biochar. For example, phenolic compounds and PAHs have been observed to contribute to the reduction in N_2O release from agricultural soils (Wang *et al.* 2013a). However, Alburquerque *et al.* (2015) demonstrated that this hypothesis was unfounded, since the presence of PAHs at typical biochar concentrations did stimulate, rather than inhibit, N_2O emissions. Moreover, even if PAH containing biochars would reduce N_2O emissions, they will definitely never be used in soils under any countries' soil and fertilizer regulations. In addition, numerous studies used clean biochars with hardly detectable traces of even the most abundant PAH (naphthalene) and these studies still showed reduced N_2O emissions (study compilations in Cayuela *et al.* (2014) and van Zwieten *et al.* (2015)).

Several studies pointed out that microbial or physical or plant immobilization of NO_3^- in soil following biochar addition could significantly contribute to the reduction of soil N_2O emissions (compilation of N_2O-biochar studies in van Zwieten *et al.* 2015). This hypothesis is reinforced by recent research showing that biochar is able to capture considerable amounts of nitrate, which is only partly detectable with standard methods and largely protected against leaching (Kammann *et al.* 2015; Haider *et al.* 2016). Nitrate capture may physically separate nitrate from denitrifiers and thus reduce nitrate availability.

On the other hand, biochar might interact with the denitrification process by modifying the ratio of denitrification products (N_2O/N_2). Thus, biochar may decrease the N_2O/N_2 ratio (Cayuela *et al.* 2013; Harter *et al.* 2014; Obia *et al.* 2015), but many questions arise from this finding. For instance, Obia *et al.* (2015) related this phenomenon to the alkalinizing effect of biochar in soil. However, the decrease in the N_2O/N_2 ratio has also been found in alkaline soils (Cayuela *et al.* 2013), where an increase in pH did not occur after biochar amendment. Harter *et al.* (2016) found that biochar addition led to the development of functional traits capable of N_2O reduction, containing typical and atypical nosZ genes. Following a different line of research, several recent articles highlight the importance of biochar redox properties, which may have a bigger impact on soil biogeochemical processes than previously thought (Prévoteau *et al.* 2016). In this line, Quin *et al.* (2015) measured N_2O reduction by injecting ^{15}N-N_2O in sterilized soil columns and demonstrated that biochar took part in abiotic redox reactions reducing N_2O to dinitrogen (N_2), in addition to adsorption of N_2O. Despite the current knowledge about the impact of biochar on denitrification, additional studies are highly needed to explore the detailed response mechanisms of denitrifiers to biochar amendment.

1.2.2. What do we know about the impact of biochar on N_2O from nitrification and other processes?

It has been described that gross nitrification rates could be increased after biochar amendment because of higher substrate availability for nitrifying bacteria (Nelissen *et al.* 2012), and several studies have analysed the impact of biochar on gross and net nitrification (Prommer *et al.* 2014). There is also a potential that biochar addition may increase nitrification (and with it, N_2O formation via nitrification pathways; Figure 1 in systems (such as needle-rich raw humus soils) due to the sorption of phenolic compounds; the latter can block or reduce nitrification. A significant increase in nitrification was seen in boreal forests after biochar addition (DeLuca *et al.* 2006; Ball *et al.* 2010) where sorption of phenols on biochar was responsible for increased nitrification rates. However, only a couple of studies distinguished among N_2O emissions from nitrification pathways (via ammonia oxidation or nitrifier denitrification) and other sources by using isotopic signatures of N_2O, inhibition techniques or molecular methods. In a laboratory incubation, Sánchez-García *et al.* (2014) found that the addition of biochar increased N_2O emissions from a calcareous soil and concluded that the N_2O formation pathway operating in the soil was nitrification (probably nitrifier-denitrification). In another study, Wells and Baggs (2014) showed that the biochar influence came primarily via ammonia oxidation, not N_2O reduction or production by denitrifiers, and increased N_2O emissions by 27%.

Dedicated studies of biochar effects on other soil sources of N_2O are largely missing. At low soil pH values, it has been observed that fungi produced N_2O instead of N_2 through codenitrification in presence of other nitrogen compounds, such as azide, salicylhydroxamic acid, nitrite and ammonium (Liiri *et al.* 2002). Since biochar can contain azide as well as other compounds, biochar

additions could theoretically enhance these fungal codenitrification processes. There are limited studies examining the functionality of the N_2O suppression through use of selective microbial inhibitors, and data did support the role of a particular microbial group in the N_2O suppression (bacteria or fungal, Lin *et al.* 2014). However, a soil pH increase caused by biochar addition could also reduce N_2O production from fungal codenitrification, thus the net outcome is unknown. Microbial community composition will likely play a role: Using an identical biochar in laboratory incubations across a series of 10 different soils, Thomazini *et al.* (2015) observed a trend for the biochar suppression that could be correlated to the total soil microbial biomass in the original soil. The knowledge of effects of biochar additions on various microbial sources of N_2O are still little understood and partly contradictory. Clearly, more research is needed to be able to design biochars for the purpose of N_2O emission reduction not only in soils, but also when using biochar in the management of N-rich agricultural (fertilizer) materials such as manures or composts (see sections 3.1 and 3.3).

1.2.3. Long-term effects of biochar addition and in old charcoal-rich soils: what do we know?

It is also still unclear how long N_2O emission reductions may persist following biochar addition to soil; or if old, black-carbon rich soils that undergo a change in their physico-chemical properties (such as Amazonian or African Dark Earths) will have a lower or higher potential for mitigating N_2O emissions compared to soils without biochar. While a lab study reported that aged biochar particles increased N_2O emissions (Spokas 2013), the opposite was observed in an experiment with >100 year-old charcoal particles from a kiln site (Kömpf 2013). Hagemann *et al.* (2016) reported that biochar still significantly suppressed N_2O emissions in the third season in the field compared to the corresponding control field site without biochar. More data are slowly emerging on old charcoal-rich soils (e.g. from historic kiln sites, Borchard *et al.* 2014a; Hardy *et al.* 2016, 2017), and more Dark Earth sites besides those in the Amazon basin have now been identified (e.g. in Liberia and Ghana, Solomon *et al.* 2016). However, to our knowledge no experiments on N_2O emissions and soil N transformations have yet been carried out on these long-term analogues compared to their adjacent native, non-black-carbon soils. For the overall question if using biochar does offer long-term benefits regarding N_2O emission suppression, exploring long-term effects is likely of great importance, since reducing peak emissions in the first years will only be a small part over the long-term. Particularly, these longer- and long-term effects are completely underexplored, and deserve much more research attention in the near future.

2. Effects of biochar application on soil CH_4 fluxes

2.1. Mechanisms of CH_4 fluxes: production and consumption in soils and net release to the atmosphere

The two biotic processes that determine the net methane (CH_4) exchange between soils/ecosystems and the atmosphere are methane production by strictly anaerobic methanogenic Archaea (Methanogens) and methane consumption by methanotrophic bacteria (Methanothrophs). Methane production takes place in all anoxic environments where organic carbon is microbially degraded (Conrad 2007a, 2007b; Whalen 2005), for example in peatlands, lake sediments, flooded rice fields, in landfills, in the guts of ruminant animals, termites or Scarabaeidae larvae (Hackstein, Stumm 1994; Kammann *et al.* 2009). Methanogens derive their energy from H_2 and carbon dioxide (CO_2) or acetate, formate, methanol or other primary and secondary alcohols and methylated compounds (Brasseur, Chatfield 1991; Conrad 1999). Methanogenesis is thermodynamically the least efficient process i.e. other reduction processes outcompete CH_4 production, if the concentration of alternative electron acceptors, such as nitrate (NO_3^-), sulphate (SO_4^{2-}), iron (Fe(III)) and manganese (Mn(IV)), is high in relation to the input of organic substrates (Lovley, Phillips 1987; Oremland 1988; Conrad 1989). Spatial and temporal variation in CH_4 can be large (e.g. Saarnio *et al.* 1997; Juutinen *et al.* 2003): Spatial variation in CH_4 flux within different microsites of the same ecosystem (e.g. Saarnio *et al.* 1997) and between different ecosystem types (e.g. Saarnio *et al.* 2009). Water table and temperature are dominant controls on CH_4 efflux in bogs and swamps whereas the effect of aquatic vascular plants (aerenchyma "ventilation") was the most important in fens or rice paddies (Turetsky *et al.* 2014). Besides the aerenchyma transport, CH_4 can also be transported with the transpiration water stream in swamp tree species as shown by Terazawa *et al.* (2007). Many $^{14}CO_2$ experiments have shown that recently fixed C is rapidly delivered from plants to methanogens (e.g. Megonigal *et al.* 1999) but the amount of exuded carbon is many times lower than that delivered via litter formation (Saarnio *et al.* 2004).

Methane consumption in soils is also ubiquitous in all terrestrial environments (Hütsch 2001; Seiler *et al.* 1984), and human land-use changes such as deforestation, ploughing and N fertilization reduce the soil CH_4 sink (Powlson *et al.* 1997). Net CH_4 consumption is due to the activity of methanotrophic α- and γ-proteobacteria. Most methanotrophs use CH_4 as the sole carbon source and need oxygen to be active (Conrad 2007a). In upland soils, methane oxidation is largely determined by the soil diffusivity for CH_4 and O_2 (Castro *et al.* 1994, 1995). According to their CH_4-oxidation kinetics, categories of "high-affinity" and "low-affinity" methanotrophs are often defined (Dunfield 2007). The first group occurs

dominantly in upland soils and can consume atmospheric and sub-atmospheric CH_4 concentrations (<1800 ppb), while low-affinity groups are found in anoxic environments (e.g. rice paddy soils) in the aerobic centimeters or millimeters of topsoil or in the oxygenated plant rhizospheres; these methanotrophs need higher CH_4 concentrations. CH_4 consumption can provide a "biofilter function" for environments with high CH_4 production (rice paddies, landfill cover soils etc.).

2.2. Biochar effects on CH_4 production and release in net CH_4-source soils

The interactions between biochar application to soil and CH_4 fluxes are not well understood, with disparate literature results (Jeffery *et al.* 2016; Song *et al.* 2016). Biochar application to paddy or flooded soils have been shown to increase (Yu *et al.* 2013; Zhang *et al.* 2012), decrease (Feng *et al.* 2012; Khan *et al.* 2013b; Lin *et al.* 2015; Qian *et al.* 2014), or have no significant effect on CH_4 emissions (Xie *et al.* 2013). In anaerobic environments, the labile C pool of biochar may theoretically function as methanogenic substrate, promoting CH_4 production (Zhang *et al.* 2010). However, the labile C pool of root exudates and root litter is by far larger, thus labile biochar C may only play a role (i) initially, (ii) when the biochar has been produced at low temperature (i.e., greater labile C fraction), (iii) in bare/fallow soils without root carbon supply, and (iv) when the biochar amount added is great (>40 t/ha) (Saarnio 2016). As an example, Zhang *et al.* (2012) did not observe increased soil CO_2 efflux over two consecutive rice cropping years with 10–40 t ha^{-1} biochar amendments, but significantly reduced N_2O emissions at increased CH_4 emissions; thus labile biochar-C is an unlikely explanation. Biochar was reported to also promote methanotrophic CH_4 consumption at oxic/anoxic interfaces in anoxic environments. This lowered the net CH_4 emissions by the "biofilter" function of bacterial (low-affinity) CH_4 oxidation before it escaped to the atmosphere. When methanotrophic organisms increasingly oxidise CH_4 in the presence of biochar at the oxic/anoxic root interface, they lower the amount of CH_4 that can enter into the plants' aerenchyma to escape (Feng *et al.* 2012).

In a greenhouse mesocosm study with sewage sludge biochar (high application rates of 5% and 10%), rice yield increased while the paddy soil turned from a net CH_4 source to a net CH_4 sink; this occurred in both rice-planted and bare paddy soil. These results may have been due to the addition of electron-accepting ash substances or nitrate with the sewage sludge biochar. In biochar-amended landfill cover soil, an increased CH_4 oxidation activity was responsible for decreasing CH_4 efflux from greater landfill depths (Sadasivam, Reddy 2015; Reddy *et al.* 2014). Here, the physico-chemical properties including air conductivity were considerably increased by biochar, i.e. the biochar effect might have been to improve the O_2 supply to methane oxidizers. A recent meta-analysis (Song *et al.* 2016) of CH_4 emissions reported that biochar application caused no pronounced change in CH_4 emissions overall but that there was significant increase in methane emissions (+19%). In another recent meta-study including papers up to December 2015, Jeffery *et al.* (2016) reported that biochar amendment to flooded and/or acidic soils had the potential to significantly reduce CH_4 emissions. These two meta-studies differ in their conclusions, which may be due to a different database and meta-analytical approach.

Biochar impacts on natural net-methanogenic environments such as salt flats and wetlands are even less well understood. Owing to the longevity of biochar and its potential mobility and migration from anthropogenic systems, it will likely migrate to coastal areas over the long-term (as recently shown for dissolved pyrogenic organic carbon, Jaffé *et al.* 2013). Lin *et al.* (2015) investigated biochar application to saline costal soils where soybean and wheat was grown. They did not find any significant effect of biochar application to such soils on the (overall low) CH_4 emissions, but they observed a yield increase of 24 and 28% in soy and wheat, respectively. In rice field studies, reductions in CH_4 and/or N_2O emissions were also often accompanied with increases in crop yields by between 10 and 20% (Dong *et al.* 2015; Khan *et al.* 2013a; Zhang *et al.* 2012), resulting in a reduced greenhouse gas intensity per kg of rice grain.

Biochar implementation may also reduce the GHG intensity per unit of agricultural product by reducing N-fertilizer and labile-C inputs at unaltered or increased yields. Qian *et al.* (2014) reported that the use of 4 different biochar-compound fertilizers made of chemical fertilizers, biochar and bentonite, at rates of well below 1 t biochar ha^{-1}, significantly improved the GHG intensity of a rice crop (by 36–56%) by: 1) the biochar-compound fertilizer increasing grain yields by 10–31%; and 2) reducing CH_4 emissions by 25–50% and N_2O emissions by 17–39%. These results coincided with a reduced overall N fertilizer input, from 210 kg N to 168 kg N ha^{-1}, and without taking the CO_2-equivalents of reduced fertilizer production and use into account (Qian *et al.* 2014). Thus, there is a considerable potential for reducing the GHG intensity of rice crop production, particularly in acidic soils (Jeffery *et al.* 2016), and that this potential extends beyond the C-sequestration potential. Large amounts of biochar do not need to be used at once since improvements were found at biochar rates <1 t ha^{-1} (Qian *et al.* 2014). Therefore, three central research topics emerge here: (1) More research is needed on mechanisms of CH_4 (and N_2O) emission reductions with biochar use from paddy and anaerobic soils including the effects on the "methanotrophic biofilter"; (2) Dedicated research on

biochar-effect-mechanism systematics to design biochars with desired properties (Thomazini *et al.* 2015); and perhaps most importantly (3) Research on biochar compound fertilizers or underfoot fertilizers (Schmidt *et al.* 2015), to achieve higher yields at reduced GHG emissions and reduced N fertilizer use.

2.3. Biochar effects on net CH_4 consumption in oxic upland soils

Well-aerated upland soils are characterised by CH_4 consumption, mediated by methanotrophic bacteria. Forest, grassland and arable land have been described as CH_4 sinks with flux rates of up to 65 μg CH_4 m^{-2} hr^{-1} (Dalal *et al.* 2008; Kern *et al.* 2012; Wang *et al.* 2005). As outlined above, in oxic/anoxic soil interfaces with a considerable CH_4 source strength, where the methane oxidiser community is dominated by low-affinity methanotrophs, significant increases in methanotrophic abundance and/or activity have been reported following biochar amendment (Feng *et al.* 2012; Reddy *et al.* 2014; Sadasivam, Reddy 2015). However, upland soils mostly host high-affinity methanotrophs, capable of consuming atmospheric methane. Species composition and biology is different to that from anoxic soils (Dunfield 2007), and the CH_4-consuming activity is quite easily hampered by human "activities" such as land conversion (particularly deforestation), N fertilization and ploughing. The number of studies using upland soils and measuring CH_4 consumption with/without biochar is currently not large. Kollah *et al.* (2015) observed significantly increased CH_4 consumption rates in a lab study with tropical soil amended with biochar (with or without organic amendments), as did Karhu *et al.* (2011) in in-field boreal, ploughed grassland soil. Karhu *et al.* (2011) assumed that the observed doubling of the CH_4 consumption in the ploughed grassland soil was due to altered gas diffusivity and water holding capacity (which increased by 11%). However, the effects may also be connected to increased N mineralization which usually occurs after ploughing, and where biochar may have prevented N (NH_4^+) inhibition by sorbing NH_4^+ (Taghizadeh-Toosi *et al.* 2012). Schimmelpfennig *et al.* (2014) observed increased CH_4 consumption in clay loam soil under laboratory condition; however, in the field this was only present in tendency. Similarly, Scheer *et al.* (2011) did not observe increased soil CH_4 consumption in a subtropical pasture that had been amended by 10 t ha^{-1} manure biochar in Australia.

In their meta-analysis, Song *et al.* (2016) reported high levels of uncertainty for CH_4 oxidation in upland soils, while Jeffery *et al.* (2016) concluded that biochar addition may reduce the CH_4 sink in neutral to alkaline upland soils. Over all data sets, biochar had a CH_4 sink-increasing (or source-decreasing) effect in soils fertilized at rates <120 kg N ha^{-1}. Translated to upland soils this indicates that, when true high-affinity methanotrophic activity is present, it may be increased by biochar application. However, when high N application rates are used this was not the case (Jeffery *et al.* 2016); with high N fertilization the CH_4-oxidising activity of an agricultural soil is often considerably reduced or completely shut down, likely due to nitrifiers replacing methanotrophs. In this case, the methanotrophic population would not be supported or improved. Taken together, the effects of biochar amendment on soil CH_4 consumption are not well understood. Here, mechanistic studies are missing in particular, and thus should be a focus for future research.

3. GHG emission reduction in animal husbandry and waste management using biochar

After nearly a decade of research where biochar was solely added to soil to assess GHG fluxes, there is a shifting perception, that biochar may also be used as a tool to achieve GHG emission reductions during the handling and management of organic nutrient-rich materials such as manures. This section focuses on the use of biochar in animal husbandry and in composting or plant-substrate production (the topic of peat replacement is addressed by Kern *et al.* 2017, this issue). In Germany, Austria and Switzerland about 90% of the traded biochar is used in animal husbandry, mainly as feed additive (in the way activated carbon is used). However, to date, this topic has been nearly neglected in biochar research.

3.1. Biochar as additive for feed and manure treatment in animal farming to reduce the emission of GHG

Charcoal has been used to treat digestive disorder in animals for several thousand years. Cato the Elder (234–149 BC) mentioned it in his classic On Agriculture: "If you have reason to fear sickness, give the oxen before they get sick the following remedy: 3 grains of salt, 3 laurel leaves, […], 3 pieces of charcoal, and 3 pints of wine." (Cato 1935, §70; O'Toole *et al.* 2016). At the end of 19th to beginning of the 20th century, charcoal was increasingly used on a regular base to increase animal performance and health (PSAC 1905; Day 1906; Savage 1917; Totusek, Beeson 1953; Volkmann 1935). Later during the last century, veterinarian research focused on activated charcoal trials mostly in the form of time-restricted medications against intoxication and bacteriological as well as viral diseases (Toth, Dou 2016; Schmidt *et al.* 2016). Only since about 2010 has biochar increasingly been used as regular feed additive in animal farming (O'Toole *et al.* 2016), usually mixed with standard feed at approximately 1% of the daily feed intake. While scientists and farmers gained most of the results and experience in cattle and chicken farming, biochar is also administered to sheep, goats, pigs, horses, rabbits, cats, dogs and extensively in fish farming (Toth,

Dou 2016; Schmidt *et al.* 2016). In a German review paper, Schmidt *et al.* (2016) evaluated more than 100 scientific papers on feeding biochar to various animal groups. Most of the studies showed for all investigated livestock species, positive (but not always significant) effects on parameters such as toxin adsorption, digestion, blood values, feed-use efficiency, cell numbers in milk and livestock weight gain; the latter may result from the pH-increasing effect of various biochars since these are mostly alkaline in nature (see 3.3). Buffering the pH in the rumen could likely prevent acidosis which is known to impact livestock weight gain. However, only a small number of researchers measured GHG emissions.

While chicken, pigs, fish and other omnivore animals provoke GHG emissions (mainly CH_4, N_2O; and NH_3) when their liquid and solid excretions decompose anaerobically, ruminants cause direct methane emissions through flatulence and burps (eructation). This is especially the case for cattle that emit daily 500 to 600 *l* gas with an average methane content between 6 and 8%. The earliest evidence that feeding of biochar might reduce cattle methane emissions came in a 2012 Vietnam study (Leng *et al.* 2012a). In-vitro studies revealed significant methane reductions of 10 to 12.7% when biochar was fed at rates between 0.5 and 1%. If biochar was blended with nitrate, methane emissions were reduced by up to 49%. Nitrate becomes a strong electron acceptor in the anaerobic rumen, keeping the hydrogen potential low, and thus replacing a function of methane producing microorganisms. Vongsamphanh *et al.* (2015) also found in in-vitro tests with 1% biochar, while using cassava in rumen fluid, a 7% reduction of methane emissions within 24 h. In-vivo trials of Leng *et al.* (2012b) revealed a reduction of enteric methane of cattle by 20% with feeding 0.6% biochar and by 40% with feeding 0.6% biochar blended to 6% potassium nitrate, leading to a highly significant animal weight increase of 25% over 98 days. This is by far the most spectacular result in reducing enteric cattle methane, but it has unfortunately not yet been supported by other in-vivo or in-vitro trials. Hansen *et al.* (2012) published the results of an in-vitro trial with various non-characterized biochars and their effect on methane production in rumen liquids. All tested biochars showed a trend to reduce methane emissions between 11% and 17%. Other groups have not repeated Leng and colleagues' promising results. The reason could be that Leng and colleagues used high temperature gasifier biochars made from rice husk, resulting usually in carbonaceous materials with high electric conductivity and electron buffering capacity (Yu *et al.* 2015) which may have had a stronger influence on the digestion reactions electrochemically than woody biochar. Using biochar feeding to reduce ruminant methane emissions is currently only an interesting perspective that needs more systematic research.

Methane adsorption capacity by biochar is typically the most investigated pathway for explaining effects when fed to animals, but adsorption cannot explain all observations. Another decisive complementary function of biochar is its electro-biochemical interaction with biological active systems, with research only recently beginning. Biochars that are produced at temperatures above 700 °C are not only good electrical conductors (Yu *et al.* 2015; Mochidzuki *et al.* 2003) but can take part in biotic and abiotic redox-reactions as an electron mediator (Husson 2013; Kluepfel *et al.* 2014; Joseph *et al.* 2015a; Liu *et al.* 2012; Shi *et al.* 2016; Van der Zee, Cervantes 2009; Yu *et al.* 2015; Kappler *et al.* 2014). A well balanced animal feed regime contains multiple electron mediating substances, however, in the high energetic diets of intensive livestock farming these compounds are often not contained in sufficient amounts (Sophal *et al.* 2013). If in these cases inert or other non-toxic electron mediators like biochar, wood vinegar or humic substances are added to the feed, many redox reactions may take place more smoothly and efficiently which could increase energy conversion efficiency and thus feed efficiency (Liu *et al.* 2012; Leng *et al.* 2013), and eventually decrease enteric and post digestive GHG production. Particularly, lowering CH_4 emissions (which are always a sign of energy loss) may be aided by the electron shuttling abilities of biochar. Moreover, it might be assumed that the buffering of the redox-potential as well as the effect of electron shuttling between various microbial species has a selective, milieu forming effect which facilitates and accelerates the formation of functional microbial consortia and syntrophic species (Kalachniuk *et al.* 1994). The latter could explain why several studies found a strong increase of Lactobacilli or a decrease of gram-negative bacteria (Naka *et al.* 2001; Choi *et al.* 2009) which seems to improve animal health. Thus, it may be hypothesized that direct electron transfers between different species of bacteria or microbial consortia (Chen *et al.* 2014) via a biochar mediator may aid in a more energy efficient digestion and thus higher feed efficiency and eventually result in lower GHG emissions (Leng *et al.* 2012a, 2012b).

When animals receive charcoal feed additives combined with Lactobacilli spraying (i.e. microbial milieu management in the stable), it is interesting to note that antibiotic use may be reduced and in some cases down to zero. Farmers in Germany who use this practice frequently report reduced veterinarian costs (Kammann, pers. comm.) that "pay" for the use of biochar and Lactobacilli solution. Reduced antibiotics may also reduce CH_4 emissions from ruminant husbandry. Recently, Hammer *et al.* (2016) showed that application of broad-spectrum antibiotics enhanced CH_4 emissions from cattle manure, and altered the gut microflora from dung beetles feeding on the manure from cows treated with broad-spectrum antibiotics. As Choi *et al.* (2009) and Islam *et al.* (2014)

showed that feeding 0.3 to 1% biochar could replace antibiotic treatment in chicken and ducks, respectively, feeding biochar plus administering Lactobacilli could have an indirect effect on GHG emissions when it is able to replace regular antibiotic "feeding". Furthermore, Joseph *et al.* (2015b) demonstrated that feeding biochar to grazing cows had positive secondary effects on soil fertility and fertilizer efficiency, reducing mineral N-fertilizing requirements which could be construed as another indirect biochar GHG mitigation effect. Thus, enabling farmers to stop or reduce administering antibiotics by using biochar and Lactobacilli may be promising, not only for animal health, but also for reducing methane emissions from animal husbandry operations (Hammer *et al.* 2016).

3.2. Calculating CO_2-equivalent balances of biochar use in animal husbandry: first considerations

Besides the possible effects of biochar feeding on ruminant CH_4 emissions, it is not unlikely that microbial decomposition of manure containing digested biochar produces less ammonia, less CH_4 and thus retains more nitrogen. This has been observed between manure composted with and without biochar (section 3.3; e.g., Sonoki *et al.* 2013; Steiner *et al.* 2010; Troy *et al.* 2013; Wang *et al.* 2013b) and may also occur when biochar is used as bedding or manure treatment additive. Ghezzehei *et al.* (2014) estimated that using biochar for liquid manure treatment could save 57,000 t NH_4 and 4,600 t P_2O_5 fertilizer per year in California alone, though this estimate is only based on laboratory adsorption tests and not on field trials. However, it cannot be excluded that digested biochar will not have the same effect on microbial decomposition, GHG emissions and plant nutrient retention as when production-fresh biochar is applied to the bedding or manure pit. To our knowledge, there are no published data on GHG-emissions in animal housing and of manure pits after feeding animals with biochar.

Easier to calculate is the C-sequestration potential of biochar that is first fed to livestock and eventually applied to soil with the manure. Assuming an average C-content of fed biochar of 80%, as required by the EBC feed certificate (EBC 2012) and produced at recommended temperatures above 500 °C resulting in H/C_{org} ratios below 0.4, at least 56% of the dry weight of the fed and manure-applied biochar will persist as stable carbon in soil for at least 100 years (Lehmann *et al.* 2015). If the global livestock would, just theoretically and for the sake of a "back-of-the envelope" assessment to explore magnitudes, receive 1% of their feed in form of such a biochar, about 400 Mio. t of CO_2eq or 1.2% of the global CO_2 emissions could be compensated (Table 1).

While the feeding of "vegetal carbon" (biochar) is permitted in the EU (EU 2011), it certainly cannot be recommended yet as in a generalized biochar-livestock feeding regime, since feed-grade certification of biochar is currently not established in most countries and since long-term effects are not sufficiently investigated. However, the potential for improving animal health and nutrient efficiency, for reducing enteric methane emissions as well as GHG emissions from manure management, and for sequestering carbon while improving soil fertility improvements, calls for increasing the scientific effort to investigate, measure and optimize the GHG reduction potential of biochar use in animal farming systems. The use of biochar in animal husbandry is one of the largest unexplored research topics within the biochar research realm. Although many unknowns and open questions exist, biochar use in animal operations appears promising from a GHG reduction standpoint and thus future research could focus efforts towards this area.

Table 1. Carbon sequestration potential of biochar fed to livestock (globally) with subsequent manure application to soil. Numbers of total livestock follow FAO statistics obtained for 2014 (FAO 2016). An intake dosage of 1% of the daily feed weight was assumed. The C sequestration potential was calculated with the assumption of 80% C in biochar, and 70% C persistence over 100 years (Lehmann *et al.* 2015; Camps-Arbestain *et al.* 2015)

Animal	Global number of individual animals	Daily intake dosage* (g bc animal^{-1} d^{-1})	Annual intake dosage (kg bc animal^{-1} yr^{-1})	Total biochar (Mio. t yr^{-1})	Total C seq. (Mio. t yr^{-1})	Total CO_2eq (Mio. t yr^{-1})
Cattle	1,482,144,415	120	43.8	64.9	36.4	133.3
Buffaloes	195,098,316	120	43.8	8.5	4.8	17.5
Sheep	1,209,908,142	50	18.25	22.1	12.4	45.3
Goats	1,006,785,725	50	18.25	18.4	10.3	37.7
Pigs	986,648,755	80	29.2	28.8	16.1	59.2
Horses	58,913,957	120	43.8	2.6	1.4	5.3
Chickens	21,321,834,000	6	2.19	46.7	26.1	95.9
Turkeys	461,453,000	6	2.19	1.0	0.6	2.1
Total				**193.0**	**108.1**	**396.3**

3.3. Biochar as composting additive

Composting is the aerobic biotic oxidation of organic residue, manure and waste for producing organic fertilizer and occasionally waste management. Labile organic carbon is transformed into humic like substances and CO_2, with N_2O and CH_4 as non-desired by-products. Emission rates of N_2O and CH_4 depend on compost management, and increase with increasing degrees of anoxia during composting. During a perfect aerobic quality composting procedure, N_2O and CH_4 emissions are usually low (Amlinger *et al.* 2003, 2008; Bernal *et al.* 2009). However, aerobic conditions cannot always be perfectly maintained, particularly when manure-rich waste is composted; here larger N_2O and CH_4 emissions are common (Li *et al.* 2016; Wang *et al.* 2013b). Thus, it is desirable to develop strategies to reduce GHG emissions during composting, particularly of nutrient-rich wet materials.

With regard to biochar, "co-composting" refers to the addition of biochar to the initial composting feedstock. During co-composting, biochar can sorb compost liquids rich in nutrients, particularly nitrate (Prost *et al.* 2013; Kammann *et al.* 2015). Furthermore, some studies report an accelerated thermophilic phase with higher compost temperatures (Kammann *et al.* 2016; Vandecasteele *et al.* 2016). CO_2 emissions are an obligate result of composting due to the decomposition of labile organic matter. This loss during composting ranges from ~40% (Vandecasteele *et al.* 2016) to ~80% (Sánchez-García *et al.* 2015) depending on the compost feedstock and composting conditions. Some studies show no effect of biochar on CO_2 emissions or increases of emission rates (Steiner *et al.* 2011; López-Cano *et al.* 2016). However, Malinska *et al.* (2014) and Vandecasteele *et al.* (2016) reported a reduction of CO_2 emissions. Vandecasteele *et al.* (2016) argue that pristine biochar used in their experiments might have physically adsorbed CO_2 (Creamer *et al.* 2014; Fornes *et al.* 2015) and thus reduced CO_2 emissions, although actual CO_2 production probably increased (e.g., increase of decomposition, higher temperatures). However, a faster rate of C loss does not necessarily mean that, overall, biochar will reduce the long-term storage of non-pyrolyzed feedstock in soils; it just means that a process that happens otherwise over longer periods is accelerated.

CH_4 and N_2O emissions from compost are non-desired side effects and can be reduced by adequate management of the composting process with regard to oxygen supply, including windrows not exceeding certain sizes, optimized water content and forced or mechanical aeration (Fukumoto *et al.* 2003; Amlinger *et al.* 2008). However, these management strategies can be costly. Recent studies indicate that biochar addition during the composting process can indeed reduce emissions of both CH_4 and N_2O probably due to enhanced access of oxygen mediated by biochar. Methane emissions are often drastically reduced by biochar addition, with reported reduction rates of 55% for chicken manure compost with biochar added at 20% w/w (Jia *et al.* 2016), >70% for organic waste compost with biochar added at 10% rate (Sonoki *et al.* 2011), and >80% for municipal solid waste compost with biochar added at 10% w/w (Vandecasteele *et al.* 2016). However, at low biochar additions (e.g. 3%), Sánchez-García *et al.* (2015) observed no significant effect on CH_4 emissions during the composting process, suggesting that a certain biochar rate during composting is necessary to reach desired reductions in CH_4 emissions.

Gaseous N losses also often decrease when biochar is used as an additive during the composting process since the pH of composts does usually stay below or around

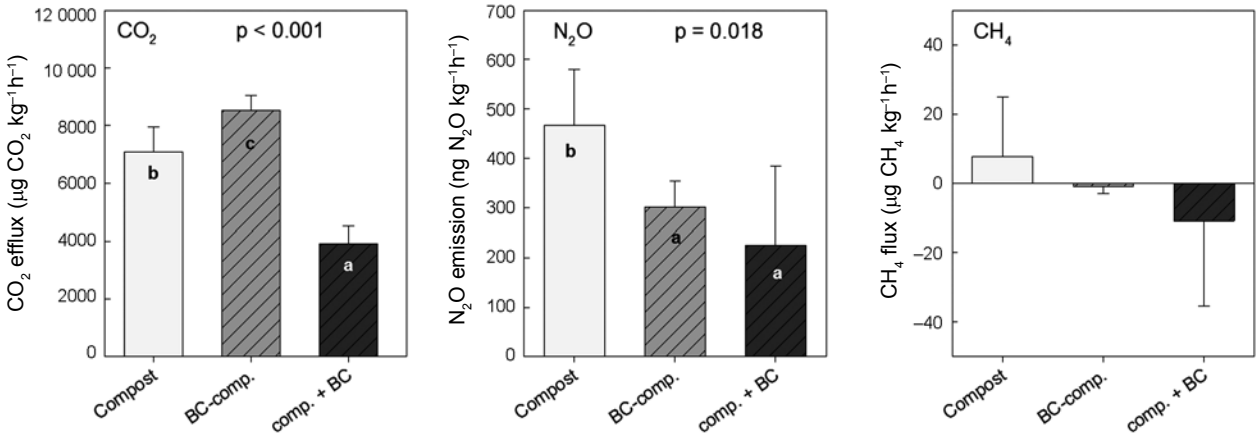

Fig. 2. Mean GHG flux rates of compost (comp), biochar-compost (BC-comp) and compost with later addition of fresh biochar (comp + BC), n = 5 + standard deviation; GHG flux measurements with 400 g substrate per 1-L Weck® jar as described in Kammann *et al.* (2012); Material properties and composting procedure described in Schmidt *et al.* (2014) and Kammann *et al.* (2015). Substrates were adjusted to 60% of their respective water-holding capacity one week before measurement. Letters indicate significant differences by one-way ANOVA (n = 5, $p < 0.05$)

pH 7.5. However, care has to be taken to prevent a strong pH increase due to (large amounts of) biochar addition in composting, with animal bedding, manure treatment or elsewhere. If values above a pH of 8 are reached, NH_3 losses may result which was systematically investigated by Chen *et al.* (2013) with biochar additions up to 20% by weight to bauxite processing sands in a pH-adjusted set-up of pH values ranging from 5 to 9. The authors reported that at low and high pH values, NH_3 losses were increased by biochar additions (at low pH when the addition of biochar was sufficiently large to provoke a pH increase), while at a high pH NH_3 losses were high anyway. However at a more neutral pH, the NH_4^+ adsorption capacity of biochar dominated which decreased N losses (Chen *et al.* 2013). However, the first available results are contradictory: While Schimmelpfennig *et al.* (2014) found a reduction of NH_3 emissions when slurry was added to a loam soil (pH 6.0) amended with biochar when compared to the same soil amended with (less alkaline) straw, Subedi *et al.* (2015) reported an increase in the NH_3 emissions from slurry when both, an alkaline biochar and an acidic hydrochar were added to the slurry. It is possible that the overall "evaporation surface" which may increase by adding a bulky material to soil or slurry may also play a role for increasing NH_3 losses. Nevertheless, the biochar-dose-to-pH-increase relationship should always be taken into account.

Studies conducted with relatively low dosages of biochar (e.g. 3–4%) showed either a reduction in N_2O emission (Wang *et al.* 2013a; Li *et al.* 2016) or no effect (Sánchez-García *et al.* 2015; López-Cano *et al.* 2016). When reductions in N_2O emissions were observed, they were pronounced but happened only during a portion of the composting process. For example, Li *et al.* (2016) reported that 3% biochar addition during composting reduced N_2O emission by 54%, which was entirely attributable to N_2O-peak suppression for only one of the eight measurement dates. This effect was further attributed to a marked reduction in the abundance of the nirK gene of denitrifying bacteria when biochar was co-composted (Li *et al.* 2016).

Ammonia (NH_3) is not a greenhouse gas, but a relevant atmospheric precursor of N_2O (shown in section 1.2), and its volatilization during composting is a relevant pathway for N loss. Biochar amendment was shown to reduce NH_3 volatilization particularly in N-rich materials such as sludge or manures, probably due to NH_4^+ sorption (Chen *et al.* 2010; Hua *et al.* 2009; Steiner *et al.* 2010; Malinska *et al.* 2014). Increased N content in the compost, in the form of NH_4^+, is a desired property for compost use as an organic fertilizer. When applied to soil, the larger quantity of N and labile C retained in the composted biochar particles (or in the biochar-compost product) may theoretically lead to higher N_2O emissions as compared to pure, non-composted biochar (Prost *et al.* 2013; Borchard

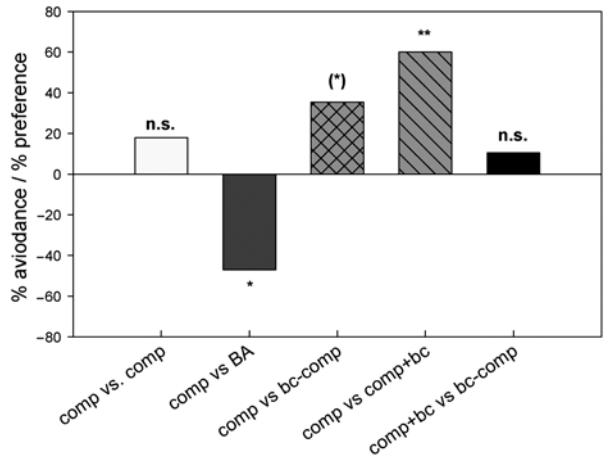

Fig. 3. Results of earthworm avoidance tests (method: ISO 17512-1:2008; Busch *et al.* 2012) with the compost (comp), biochar-compost (bc-comp) and compost with later addition of fresh biochar (comp+bc), material properties and composting described in Schmidt *et al.* (2014) and Kammann *et al.* (2015). "Comp vs. comp" is the control and did not result in an effect (as needed); boric acid (BA) was the "positive control" where avoidance behaviour confirmed that the test was valid. Bars give means of n = 5 repetitions per treatment (50 worms), shown as preference or avoidance of the respective substrate (Fishers exact test, (*) $p < 0.1$, * $p < 0.05$, ** $p < 0.01$, n.s. not significant)

et al. 2014b). This was, however, not observed in a planting study using co-composted (nitrate-enriched) biochar as soil amendment (Kammann *et al.* 2015), or when the compost itself was tested (Fig. 2).

At the moment, results are just snapshots of first composting experiments with biochar, and studies include a wide range of experiments from 150 g mixtures in jars (Jia *et al.* 2016), 45 L composting reactors (Malinska *et al.* 2014), 1 m³ gardening compost boxes (Prost *et al.* 2013) to 60×3×2 m windrows with forced aeration and automated mechanical turning (Vandecasteele *et al.* 2016) or daily machine turning (Schmidt *et al.* 2014; Kammann *et al.* 2015) at varying time scales (few weeks to several months). Generally, the value of composting studies would be increased if the resulting compost quality is subsequently evaluated by plant germination and growth tests (e.g. Hua *et al.* 2012, soil faunal tests, e.g. Fig. 3) and field application trials.

Biochar is a promising tool to optimize composting, improve compost quality and charge biochar with plant-available nutrients and reduce non-CO_2 GHG emissions during composting. However, the longer-term effects of biochar-composts in soils are largely unexplored. More systematic studies with different combinations of N-rich and N-poor feedstock and different well-characterized biochars under comparable, praxis-relevant conditions are needed, and subsequently product quality and its effects on soil GHG emissions after soil amendment should be investigated.

4. Novel approaches of using biochar for GHG emission reductions in agriculture

GHG emission reductions with biochar use may not necessarily be direct; rather, reductions may result from alternative waste stream management involving pyrolysis. Another option is to use biochar as a fertilizer carrier to match plant demands more closely, enabling the reduced use of common mineral fertilizers and hence associated N_2O emissions (see 3.1, Qian *et al.* 2014). This section addresses novel biochar concepts for reducing the overall GHG intensity of agricultural production, including those from unwanted NO_3^- and NH_3 losses and deposition, causing N_2O emissions elsewhere (Turner *et al.* 2015).

4.1. Biochar as carrier for (organic) underfoot fertilizers

The use of large biochar amounts (e.g. 10–20 tons) being ploughed into soils does usually not increase crop yields sufficiently to justify the costs (Jeffery *et al.* 2015; Biederman, Harpole 2013; Ruysschaert *et al.* 2016). Here, the economy of biochar use may be better for special crops as long as yield increases without negative effects on crop quality can be realized, as observed by Baronti *et al.* (2014) and Genesio *et al.* (2015) for grapevine cultures. However, grapevines are low-N cultures with low N_2O emissions (Marras *et al.* 2015) compared to higher N-fertilizer demanding vegetable crops such as radish, carrot, spinach or potato. These receive much higher N fertilizer amounts, often with several cropping cycles per year, resulting in high N_2O emissions (Ruser *et al.* 1998; Min *et al.* 2016). In highly fertilized vegetable crops, biochar amendment may play a role in significantly reducing the greenhouse gas intensity per unit of crop, likely by reducing N_2O emissions and partly by yield increases (Jia *et al.* 2012; Li *et al.* 2015). However, use of biochar in these situations, as with others, comes with a cost.

In order to reduce costs, it is desirable to maximize the effect of biochar per unit applied, suggesting that optimum application rates might be fairly low especially if the product can be concentrated around the root zone. Indeed, root zone application of only 2–4 t ha^{-1}, together with fertilizer in conservation farming systems, has been reported to improve yields in acidic sandy soils (Cornelissen *et al.* 2013). In addition, using biochar as an ingredient of, or carrier matrix for, mineral or organic fertilizer blends ("fertichars") has recently been investigated as a promising strategy to reduce the need for large biochar amounts while improving crop yields and/or reduce N use (Joseph *et al.* 2013). Supporting this contention, Qian *et al.* (2014) showed that biochar organo-mineral fertilizer blends reduced the GHG-intensity of rice crop production even at reduced N fertilization rates and at very low amounts of biochar (<1 t ha^{-1}).

In practice, the on-farm availability of clean, inexpensive, and amounts required will decide whether biochar will be used, particularly in rural areas in developing countries (Cornelissen *et al.* 2013). In Nepal, a self-made organic "fertichar" was used as underfoot fertilizer in 8 different farmer sites (fertile loamy soils) and compared to the same nutrient additions without biochar. Here, pumpkin yields were consistently increased by 400% with the urine-loaded biochar applied as underfoot fertilizer, and multiple field trials on different soils, crops and biochar consistently showed a growth-promoting effect of the organic biochar-root zone fertilizer (Schmidt *et al.* 2015). It has not yet been tested if these biochar-fertilizer blends will reduce GHG emissions compared to an equal or greater amount of standard fertilizer. However, we argue that GHG emission reductions (namely those of N_2O) per unit of crop yield may be achieved for the following reasons: (1) The fertichar blends mean a comparatively high amount of biochar concentrated together with the nutrients, i.e. within the "biochar concentration range" found to reduce N_2O emissions in soils considerably in meta-studies (Cayuela *et al.* 2014); (2) By stimulating plant growth, N uptake increases, which reduces the availability of N for nitrification / denitrification and hence N_2O formation; (3) When applied underfoot, the total land area that receives N fertilizer and is able to produce N_2O emissions is smaller; (4) By using root-zone fertichars, the overall amount of N fertilizer that needs to be applied, per unit of crop produced, can likely be reduced (Joseph *et al.* 2013; Qian *et al.* 2014). This may automatically reduce the GHG emissions (as a percentage of the applied fertilizer); and (5) Biochar will reduce the soil bulk density (e.g. Obia *et al.* 2016) and increase soil air capacity, which has been shown to reduce N_2O production due to improved oxygen supply in upland soils. However, it is simply unknown if this type of application will reduce the GHG intensity of crop production and if this will occur on different soils and climatic zones alike. Further research is needed on fertilizer-biochars, either as broadcast granules (Qian *et al.* 2014) or as underfoot fertilizers (Schmidt *et al.* 2015), since these techniques may offer a win-win in terms of reducing GHG emissions concomitantly with improved yields.

4.2. Pyrolysis as alternative waste stream management technique

Over the past twenty years, localization of confined animal feeding operations (CAFOs) in the USA has brought about a massive production of animal manures (Gollehon *et al.* 2001). Traditional management practices have applied manure to crops as a fertilizer, yet long-term manure applications have caused nutrient imbalances in soils far in excess of what crops can assimilate (Barker, Zublena

1995; Kellogg *et al.* 2000). The situation is so severe that CAFO producers must have nutrient management plans (e.g., MDE 2016; ISDA 2016) or manure application can be restricted if soils contain excess N and/or P. A clear need exists for alternate animal waste management and recycling methods.

Thermal processing of animal manures using gasification and pyrolysis technology is gaining considerable interest as an alternate treatment option because of the energy quantity generated (Cantrell *et al.* 2007, 2008). Manure gasification is a popular conversion process, yet the higher temperatures (>900 °C) required for conversion into gases, along with impurities in the feedstocks (e.g. salts, silicates, etc.), oftentimes limits the conversion efficiency (Lv *et al.* 2010) and can corrode downstream metal surfaces (Demirbas 2005). Thus, pyrolysis is more often regarded as the effective method of processing biomass to produce a combination of non-condensable gases, bio-oil, and biochar (Antal, Grønli 2003). Manures can be pyrolyzed at various temperatures (e.g. 300 to 750 °C) to create biochars. Afterwards, the energy content (as MJ/kg) contained within biochar identifies the quantity of energy generated per equivalent dry weight and serves as a convenient energy index relative to coal (Table 2). Globally, animal waste pyrolysis is performed to create a thermal energy source, to generate heat for animal confinement stables, for the production of bio-oil, and to produce biochar, a nutrient-enriched end-product to be used as a soil amendment (Laird *et al.* 2009; Lee *et al.* 2013) or fertilizer replacement.

Table 2. Net thermal energy in various manures, and their biochars pyrolyzed at various temperatures, as compared to hard/soft coals, gasoline and methane. na = not available

Feedstock	Thermal temperature (°C)	Energy content (MJ/kg)	Source
Poultry litter	0	15	Novak *et al.* (2013)
	700	14.2	Novak *et al.* (2013)
Dairy manure	0	17.6	Cantrell *et al.* (2012)
	350–700	19.0–20.9	Cantrell *et al.* (2012)
Swine Solids	0	19.4	Cantrell *et al.* (2012)
	350–700	15.1–21.1	Cantrell *et al.* (2012)
Human Feces	300–750	13.8–25.6	Ward *et al.* (2014)
Hard/soft coal	na	29.3–33.5	Euronuclear (2016)
Gasoline	na	43–47	Energynumbers (2005)
Methane	na	55.5	Energynumbers (2005)

Following manure pyrolysis, it has been shown by Gaskin *et al.* (2008) that nutrient availability may be decreased in the biochar, which may make manure-based biochar use attractive in terms of land areas where nutrient management plans are necessary; biochars may be able to supply a more balanced quantity of essential plant nutrients without degradation in environmental quality due to nutrient over-application. Furthermore, an additional benefit similar to previously mentioned underfoot fertilizer-biochars, manure-based biochar may significantly reduce overall farm-management based GHG emissions simply by reducing mineral fertilizer use (Cayuela *et al.* 2014; Nguyen *et al.* 2014).

The production of animal manure biochar via pyrolysis, and the subsequent partial or complete replacement of standard mineral NPK fertilization by manure-biochar, offers the following pathways to N_2O emissions reduction: (1) Reduced N use which reduces the overall environmental burden by limiting N_2O formation both from direct manure application as well as emissions originating from off-site N pollution (e.g. NH_3 emissions, N export via overland flow, N leaching to groundwater, Turner *et al.* (2015); (2) The use of manure-based biochar may reduce N_2O emissions compared to using the same amount of N in the mineral or manure form. Cayuela *et al.* (2014) examined 107 articles related to manure-based biochar land application, showing a –46 to +39% change in N_2O emissions, with average N_2O emission changes close to 0%. Thus, changes or reductions in N_2O emissions is likely process dependent, something that was not examined by Cayuela *et al.* (2014). However, Subedi *et al.* (2016) added poultry litter or swine manure biochar to two different soils. The authors showed that poultry litter or swine manure biochars pyrolyzed at 400 °C produced the same N_2O emission factor (i.e., N_2O emitted as a percentage of the total N supplied) as control soils. When pyrolyzed at 600 °C, the N_2O emission factor decreased for both biochars as compared to controls. Higher temperatures during production will result in manure-based biochars containing lesser quantities of easily degradable C compounds (e.g., volatile compounds) that are available for denitrifiers, leading to less likelihood of anaerobic soil conditions (e.g., Liu *et al.* 2014) and thus lower N_2O emissions. Obviously, in order to reduce N_2O emissions when applying manure-based biochars, easily degradable C sources needs to be at a minimum; and (3) The use of pyrolysis for heat generation in animal housing, and/or the use of manure biochar for energy production, can be implemented to reduce GHG emissions when it replaces the use of fossil fuels for the same purpose (see energy content comparison in Table 2).

Conclusions: promising options for GHG emission reduction and future research needs

After a decade of intense biochar research, it has become clear that biochar soil amendments are able to reduce N_2O emissions (i.e. emission peaks). Biochar can also reduce CH_4 emissions, particularly in flooded soils, and when N fertilization rates are not too high. However, great uncertainty still exists with respect to biochar use and its GHG reducing effect as associated with different biochars and soil types/conditions. This is due to the lack of understanding of mechanistic biochar effects. Good "candidate" mechanisms that might explain N_2O emission reductions are pH increases (liming effect) and changes in microbial community composition, particularly changes in the denitrifier gene expression and abundance, and N (mostly nitrate) capture in biochar particles. It is likely that these mechanisms work in concert under field conditions. Good "candidate" mechanisms that might explain CH_4 emission reductions are the stimulation of methanotrophic low-affinity communities at the anoxic/oxic interface in reduced environments as well as the electron shuttling and redox activity of biochars (e.g. rice paddies and ruminant guts).

Biochar use in animal husbandry is economically promising and the dominant route of biochar use in central Europe; however, research on GHG emission reductions is largely lacking. Studies dealing with the medical use of charcoal/biochar in animal feeding nearly never characterize char properties. Biochar shows a strong potential for reducing GHG and NH_3 emissions in the composting of wet nutrient-rich materials, particularly when composts are not so frequently turned (aerated, i.e. "lazy composting"). Yet, more detailed studies on the dynamics of this effect along the composting process are needed, as well as a cost-benefit evaluation for potential implementation.

One new promising option of using biochar for improving the GHG-intensity (yield-to-GHG emission ratio), via both increasing yields plus reduced GHG emissions, may be the use of designed (organic) "FertiChars" (biochar as fertilizer carrier) administered as concentrated root zone fertilizers. Here the biochar-to-N ratio may certainly be in the range where biochar should have an effect as well as effectively and environmentally deliver optimal nutrient concentrations to plants. However, to date, GHG flux measurements of this implementation pathway are lacking and strongly call for future research. We conclude that the use of biochar in agriculture provides a unique opportunity to reduce the non-CO_2 greenhouse gas "cost" per unit of yield produced, yet future research is required to maximize its benefits.

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Contribution

CK developed the structure, wrote sections, contributed to sections and coordinated writing. JI and NH helped streamlining and finalizing the manuscript in addition to section contributions. All other authors contributed equally by reviewing the literature within their particular field of expertise and writing sections of the manuscript. (Therefore, all authors besides authors 1–3 are listed in alphabetical order.)

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Claudia KAMMANN. PhD, Professor for Climate Change Research for Special Crops, Institute for Soil Science and Plant Nutrition, Hochschule Geisenheim University, Geisenheim, Germany, 58 peer-reviewed articles in scientific journals, 7 book chapters, 2453 citations, h-index 27 (Google Scholar).

Jim IPPOLITO. PhD, Associate Professor in the Department of Soil and Crop Sciences, Colorado State University, Fort Collins, CO, USA. 170 publications in peer reviewed scientific publications, book chapters, conference reports, and experiment station technical reports and bulletins. 2099 citations, h-index 24 (Google Scholar).

Nikolas HAGEMANN. PhD, Nikolas Hagemann, PhD. Researcher at University of Tuebingen, Germany. 2 peer-reviewed articles, 5 book chapters, 16 citations, h-index 3 (Google Scholar).

Nils BORCHARD. PhD, Soil Science Expert at the Center for International Forestry Research (CIFOR), Jalan CIFOR, 16115 Bogor Barat, Indonesia and at the Deprtament of Soil Science and Soil Ecology, Institute of Geography, Ruhr-University of Bochum, Universitätsstr. 150, 44801 Bochum, Germany. 15 peer reviewed publications, 3 non-peer reviewed publication, 156 citations, h-index 7 (WoS).

Maria Luz CAYUELA. PhD, is a researcher at the National Research Council in Spain. She has published 40 articles in peer- reviewed journals and 6 book chapters. 2046 citations, h-index 24. (Google Scholar)

José M. ESTAVILLO. PhD, Professor of Plant Physiology, Faculty of Science and Technology, University of the Basque Country/EHU, Spain, 40 peer-reviewed articles in scientific journals, 783 citations, h-index 15 (WoS).

Teresa FUERTES-MENDIZABAL. PhD, Researcher, Faculty of Science and Technology, University of the Basque Country/EHU, Spain, 9 peer-reviewed articles in scientific journals, 71 citations, h-index 5 (WoS).

Simon JEFFERY. PhD, Senior Lecturer in Sustainable Technology. Crop and Environment Sciences Department, Harper Adams University, Newport, Shropshire, TF10 8NB. United Kingdom. 24 peer reviewed articles in scientific journals, 3 policy reports to the European Commission. Editor of two atlases (including lead editor of one), 3 book chapters. 1768 citations, h-index 15 (Google Scholar).

Jürgen KERN. PhD, Senior Scientist of Biogeochemistry, Leibniz Institute for Agricultural Engineering Potsdam-Bornim, Germany, 1211 citations, h-index 16 (Google Scholar).

Jeff NOVAK. PhD, Research Soil Scientist, in the United States Department of Agriculture, Agriculture Research Service-Coastal Plains Research Center, Florence, South Carolina, USA. 150 publications in peer reviewed scientific publications, book chapters, and conference reports. 4130 citations; h-index 26 (Scopus).

Daniel RASSE. PhD, Senior Scientist and Head of the Department for Soil Quality and Climate Change at the Norwegian Institute of Bioeconomy Research, Aas, Norway, 55 peer-reviewed articles in scientific journals, 2948 citations, h-index 25 (Scopus).

Sanna SAARNIO. PhD, Lecturer in the Department of Environmental and Biological Sciences, University of Eastern Finland, Finland, 52 peer-reviewed articles in scientific journals or books, 1400 citations, h-index 20 (WoS).

Hans-Peter SCHMIDT. Director of Ithaka Institute for Carbon Strategies, Arbaz, Switzerland, 16 peer-reviewed articles, 9 book chapters, 259 citations, h-index 9 (google scholar).

Kurt SPOKAS. PhD, Research Soil Scientist with the United States Department of Agriculture-Agricultural Research Service, Soil and Water Management Laboratory, 1991 Upper Buford Circle, St. Paul, MN 55108. He has produced over 200 scientific publications in peer reviewed journals, book chapters, conference proceedings, and bulletins. 4904 citations; h-index 31 (google scholar).

Nicole WRAGE-MÖNNIG. PhD, Professor of Grassland and Forage Sciences, Faculty of Agricultural and Environmental Sciences, University of Rostock, Germany, 45 peer-reviewed articles in scientific journals, 6 monographs and book chapters, 1968 citations, h-index 18 (WoS).