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Short communication

N_2O and CH_4 emissions from N-fertilized rice paddy soil can be mitigated by wood vinegar application at an appropriate rate

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ABSTRACT

To understand the impacts of wood vinegar (WV), a by-product of biochar production, on N_2O and CH_4 emissions and their total global warming potential (GWP_t) from N-fertilized rice paddy soil, a soil column experiment was conducted using three treatments: 240 kg urea-N ha^{-1} accompanied with 0, 5, and 10 t WV ha^{-1}, respectively. Results showed that N_2O and CH_4 emission flux patterns were dominated by water regime of rice growth cycle, which was independent with WV application. The total N_2O, CH_4 emission loads and GWP_t over rice season of three N received treatments were 6.41–8.85 kg ha^{-1}, 127.7–405.0 kg ha^{-1}, and 5.24–12.03 t CO_2-e ha^{-1}, respectively. Rice seasonal N_2O and CH_4 emissions were synchronously mitigated by 22.4% and 36.4%, respectively, when WV was applied at 5 t ha^{-1}. Consequently, 5 t ha^{-1} WV treatment reduced 31.5% of GWP_t compared with the urea treatment. In addition, 10 t ha^{-1} WV treatment exerted a more positive effect on suppressing N_2O with 27.6% reduction. However, it increased GWP_t by 57.2% because its CH_4 emission load was increased by 101.8%. In conclusion, WV amendment applied at an appropriate rate (5 t ha^{-1}) or combination with other CH_4 control technologies were suggested to reduce both N_2O and CH_4 emissions and thereby the GWP_t in N-fertilized rice paddy soil.

1. Introduction

Methane (CH_4) and nitrous oxide (N_2O) are two powerful greenhouse gases (GHGs), being with a global warming potential (GWP) of 25 and 298 times greater than carbon dioxide on a 100-year mass basis (IPCC, 2013). Flooded rice (*Oryza sativa* L.) systems simultaneously emit CH_4 and N_2O to atmosphere, due to the favorable production, consumption, and transport systems between the rice rhizospheric soil and the atmosphere, especially in the rice paddy soil which received high loads of nitrogen (N) fertilizer (Linquist et al., 2012; Ji et al., 2015; Yao et al., 2017). China, as the world's leading producer of rice, grows rice on approximately 30 million ha per year, with dramatically increased N consumption (FAO, 2012). It is estimated that 7.7–8.0 Tg CH_4 and 138–154 Gg N_2O are emitted from Chinese rice fields (Liang et al., 2013). Therefore, it is of great importance to mitigate both CH_4 and N_2O emissions from N-fertilized rice paddy soil.

Recently, biochar has been considered as a promising soil amendment, which can improve soil quality, enhance crop productivity and mitigate GHGs emissions (Jeffery et al., 2011; Zhang et al., 2012; Pratiwi and Shinogi, 2016; Feng et al., 2017; Niu et al., 2017; Sun et al., 2017). Based on these positive impacts, biochar will likely have large-scale applications in the future. However, this will lead to the great generation of by-products. Wood vinegar (WV), which is also known as pyroligneous acid, is one of the major by-products during biochar production and can be incorporated into farmland soil (Yatagai et al., 2002; Lin and Hwang, 2009; Hanger, 2013a). Therefore, it is interesting to know the impacts of WV on GHGs emissions after incorporation into farmland soils, which will expand the application scope of biochar and its by-products.

The main substances of WV include many organic chemical components (Yatagai et al., 2002; Baimark and Niamsa, 2009). Herein, 2, 6-dimethoxyphenol, 2-methoxyphenol, and 3, 5-dimethoxy-4-hyoxytoluene, were found as main three components in WV according to Yang et al. (2016). As reported in previous literature, WV soil addition has showed beneficial effects including crop productivity improvement (Kadota and Niimi, 2004; Lashari et al., 2013, 2015; Polthanee et al.,

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2015), glyphosate leaching reduction (Hagner et al., 2013b), as well as NH_3 volatilization mitigation (Win et al., 2009). However, whether WV application can mitigate CH_4 and N_2O emissions from rice paddy soil at the same time are not well documented (Ogawa and Okimori, 2010). If so, it is a theoretical and practical innovation on how to effectively mitigate the GHGs emissions from rice paddy systems. Therefore, related works should be conducted to expand the usage scopes of by-products of biochar, as well as to protect the atmospheric environment.

The organic compounds in WV will provide C source for microorganisms in soil. Previous works reported that microorganisms and microbial activity changes in agricultural soils as results of the organic contents in the added WV (Baimark and Niamsa, 2009; Hanger, 2013a; Lu et al., 2015). In addition, the acid property will change the soil environment. Overall, C source, pH change, microbial activity were the direct factors that related to the GHGs emissions from agricultural soil (Ji et al., 2015; Li et al., 2015; Niu et al., 2017; Zhao et al., 2017). We therefore hypothesize that WV addition may influence the CH_4 and N_2O emission from flooded rice paddy receiving N fertilizer. If so, to which direction and to what degree need accurate assessments.

Here, we conducted a soil column experiment with rice cultivation to evaluate the actual effects of WV amendment on GHGs emissions. In brief, the purpose of this study was to investigate the effects of WV at three application rates (0, 5 and 10 t ha^{-1}, respectively) on CH_4 and N_2O emission flux patterns, the cumulative CH_4 and N_2O emission loads, as well as the total global warming potential (GWP_t) over rice season in a paddy soil with N fertilization.

2. Materials and methods

2.1. Background information and soil column installation

The soil used for repacking to soil column (25 cm in diameter and 70 cm in height, Supplementary Fig. 1) was collected from a paddy field used for more than 50 years in Zhoutie town, Yixing city, Jiangsu Province, China (31.4765 °N and 119.9861 °E). The test soil was classified as Hydragric Anthrosol and the top layer (0–20 cm) soil has following properties: pH 6.38 (soil: water, 1: 2.5), total N 1.56 g kg^{-1}, total P 0.96 g kg^{-1}, total K 4.12 g kg^{-1}, and organic matter 2.28%. The detailed processes of soil sampling, pretreating and repacking to soil columns were performed according to our previous work (Sun et al., 2013; Feng et al., 2017). Tested WV was derived according the methods as described in Yan et al. (2011) and Feng et al. (2017). The WV was the liquid by-product when biochar is produced via pyrolysis of fruit wood waste in a continuous slow pyrolysis system at 500–600 °C. The basal properties of WV were: pH 6.31, total N 258 mg L^{-1}, total P 2.17 mg L^{-1}, total K 60 mg L^{-1}, EC 2.18 ms cm^{-1}, TOC 1.4 g L^{-1}. The main organic compounds of WV included 4-hydroxy-4-methyl-2-pentanone, acetic acid, propionic acid, *n*-Hexadecanoic acid, etc.

2.2. Experimental treatments and managements

There were three treatments with three replicates including: 1) Urea: with 240 kg N ha^{-1} but without WV; 2) Urea + WV5: with 240 kg N ha^{-1} and 5 t WV ha^{-1}; 3) Urea + WV10: with 240 kg N ha^{-1} and 10 t WV ha^{-1}.

The rice seedlings were transplanted with a density of 6 plants per soil column on July 7, 2016. Urea fertilizer supplied the N demanded and was split applied as basal fertilizer (BF) and two time supplementary fertilizers (SF1 and SF2, respectively) in ratio of 40%: 40%: 20% on July 7, July 18 and August 19, 2016, respectively. Meanwhile, 90 kg P_2O_5 ha^{-1} (in the form of calcium superphosphate) and 120 kg K_2O ha^{-1} (in the form of KCl) were both applied as BF. All tested soil columns were continuous flooded with 3–5 cm water depth (expected for one mid-season drainage period from August 5 to 13, 2016). Local farmer' practice was adopted to control the disease and pest. The rice plants were harvested on November 30, 2016.

2.3. N_2O and CH_4 measurement

GHGs samples were collected using static chambers (29 cm in diameter and 100 cm in height) according to the modified method in Sun et al. (2013). Gas samples were taken on the 1st, 3rd, 5th, and 7th day after each split urea N fertilization applied and after surface floodwater drained. During other monitoring periods, gas samples were collected two or three times per month. Before gas samples were collected, the chambers were placed on the basal fixed frame in each soil column and sealed with water. For each observation, four gas samples were extracted from the chamber with a 50 mL syringe at the 1st, 16th, 31st, and 46th min after chamber sealed and were immediately transferred into evacuated vials. The N_2O and CH_4 concentrations analysis, GHGs emission rates calculation and the rice seasonal cumulative N_2O and CH_4 emission loads evaluation were performed according to Sun et al. (2013).

2.4. Total global warming potential (GWP_t) of N_2O and CH_4

GWP_t (kg CO_2-e ha^{-1}) was used to compare the total GHGs emission in CO_2-equivalents per hectare under different treatments. Based on this method, CH_4 and N_2O emission can be evaluated and help assessing the potential trade-off effects between soil N_2O and CH_4 emissions as affected by WV amendment in rice paddy soil. In the current study, GWP_t was calculated using the following equation (Zhang et al., 2012):

$$GWP_t = 298 \times EM(N_2O) + 25 \times EM(CH_4)$$

where $EM(N_2O)$ and $EM(CH_4)$ represent rice seasonal emissions of N_2O and CH_4, respectively; 298 and 25 are coefficients for transforming N_2O and CH_4, respectively, into equivalent CO_2 emissions within a 100-year time horizon.

2.5. Data statistical analysis

One-way analysis of variance (ANOVA) was performed to assess the effects of WV application with different rates on the N_2O and CH_4 emissions from N-fertilized rice paddy soil. Statistical differences ($P < 0.05$) among tested individual treatments were determined using Duncan multiple comparison tests (SPSS Ver. 16.0 for Windows, SPSS Inc., Chicago, IL, USA).

3. Results and discussion

3.1. N_2O emission flux pattern and cumulative emission load during rice season

In rice season, similar N_2O emission flux pattern was observed in three treatments, which was mainly determined by water regime (Fig. 1). This was consistent with previous results evidenced from rice paddy soil (Zou et al., 2005; Liang et al., 2013; Sun et al., 2015). In the continuous flooded period until August 5, almost no N_2O emission was observed, even after BF and SF1 were applied. During the mid-season drainage period, a large proportion of N_2O emissions was detected immediately after disappearance of the floodwater. The N_2O emission rates of N-fertilized treatments came to peaks with 3205–4858 μg m^{-2} h^{-1} (Fig. 1). Then the soils were re-flooded, and N_2O emission rates dropped off immediately and kept at a very low level. This could be mainly attributed to the anaerobic conditions in continuous flooded paddy soil (Sun et al., 2015).

Considerable amount of N_2O emissions with 6.41–8.85 kg ha^{-1} was recorded in three treatments. In comparison with the urea treatment, WV incorporation significantly ($P < 0.05$) reduced the cumulative N_2O emissions by 22.4–27.6% (Fig. 2), though no obvious difference was found between Urea + WV5 and Urea + WV10 treatment groups. Mitigation of N_2O emission from rice paddy soils received WV is of great significance since N_2O is a powerfully potent GHG. As result of the

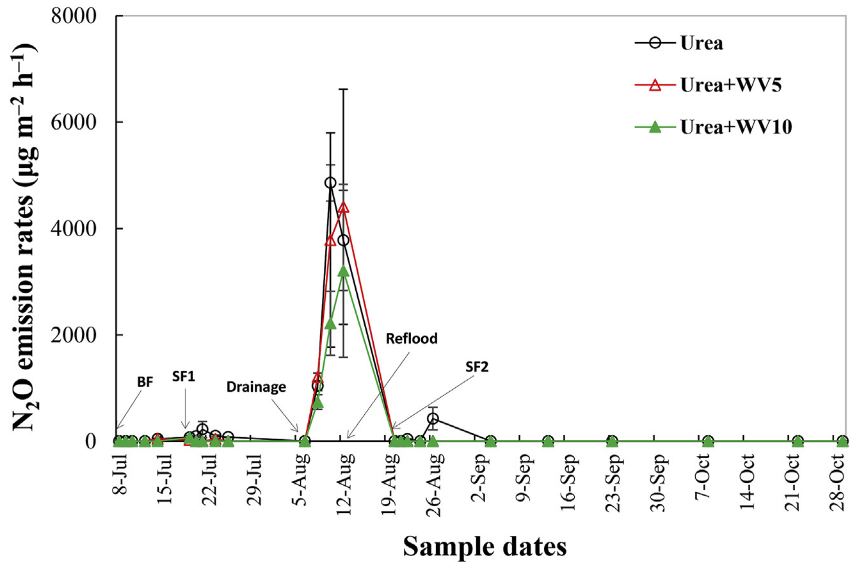

Fig. 1. Seasonal variations in N_2O emission fluxes from rice paddy soil. BF: basal fertilization; SF1: first supplementary fertilization; SF2: second supplementary fertilization. Data were presented with mean value ± SD (*n* = 3).

organic C input and soil pH change, WV amendment could potentially favor the activity of N_2O reductase from denitrifying microorganisms, while inhibited the activity of reductases involved in the conversion of NO_3^- to N_2O (Yanai et al., 2007; Ranatunga et al., 2018). Indeed, changes in soil microbial community structure and enzyme activity after WV addition have been reported (Lu et al., 2015; Yang et al., 2016). As a consequence, what is the main emission factor contributing to the reduction of N_2O from N fertilizer plus WV needs further investigations.

3.2. Methane (CH_4) emission flux pattern and cumulative emission load during rice season

Fig. 3 shows the CH_4 flux pattern from paddy soil throughout the rice growth period. There was no difference in the pattern of seasonal variations of CH_4 flux among the treatments whether WV was added or not. Seasonal pattern of CH_4 emissions varied with water regime (Zou et al., 2005; Zhang et al., 2012), which was also confirmed in our present study (Fig. 3). The CH_4 flux generally increased and reached peaks at about 30 days after rice seedlings were transplanted and then dropped off. After soil re-flooded, the CH_4 flux increased to another peaks and remained at low level afterwards, until the rice was harvested. The patterns of seasonal variations in CH_4 flux from rice paddy plots were quite different from those of N_2O flux (Figs. 2 and 3): CH_4 mainly emitted during water flooding stage, whereas N_2O emitted mainly during mid-season drainage period. Similarly, a trade-off emission pattern between CH_4 and N_2O resulting from mid-season drainage has been well documented in rice paddy soils (Zou et al., 2005; Yao et al., 2017). Based on the observed data in the present work, WV amendment did not change either the CH_4 or N_2O emission flux pattern of rice growth cycle.

Total of CH_4 emission loads of N-fertilized treatments were from 127.7 kg ha^{-1} to 405.0 kg ha^{-1} in the present study. Results in Fig. 4 shows that CH_4 emission was reduced by 36.4% when WV applied at 5 t ha^{-1}; however, it was increased dramatically by 101.8% when the WV application rate increased to 10 t ha^{-1}.

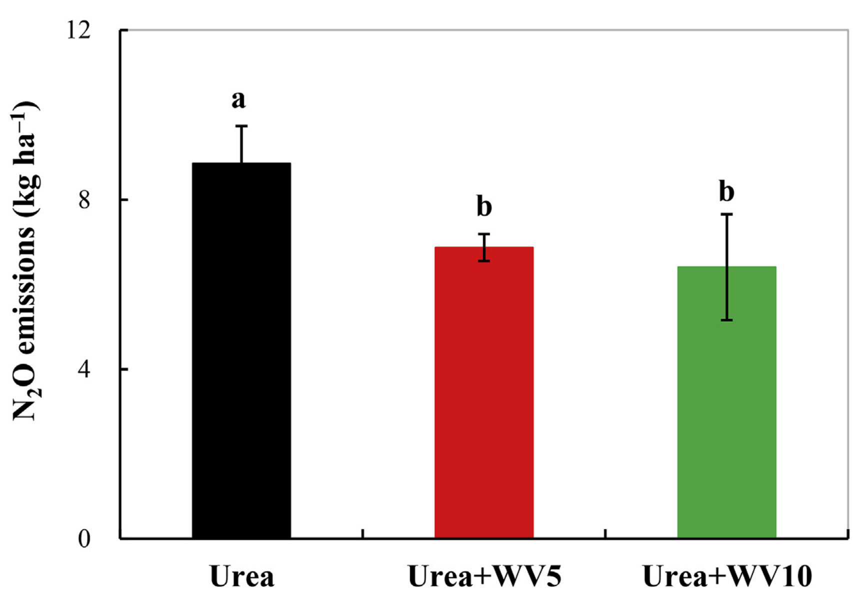

Fig. 2. Cumulative N_2O emissions from paddy soil over a single rice cycle as impacted by wood vinegar (WV) addition. The *bars* represent the standard deviation of triplicates (*n* = 3). Different *Lowercase letters* indicate significant ($P < 0.05$) differences among treatment means.

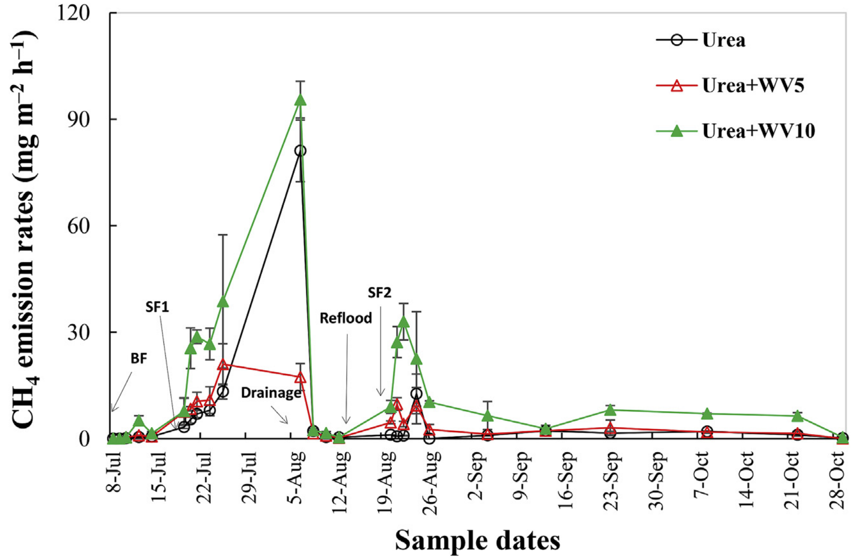

Fig. 3. Seasonal variations in CH_4 emission fluxes from rice paddy soil. BF: basal fertilization; SF1: first supplementary fertilization; SF2: second supplementary fertilization. Data were means of three triplicates ($n = 3$).

It is well known that multiple factors contribute to the intensity of CH_4 emissions from rice paddy soil (Schimel, 2000; Bridgham et al., 2013). The CH_4 emitted from rice paddy soil is primarily determined by three processes: CH_4 production, oxidation, and transport from soil to atmosphere (Cai et al., 2007; Linquist et al., 2012). WV contains many organic components and the composition is very complicated (Mum and Ku, 2010; Yang et al., 2016), also reflected from our WV. It is possible, WV enhanced CH_4 oxidation in the root rhizosphere which leads to a reduction in CH_4 emissions under Urea + WV5 treatment.

However, WV applied at higher load ($10\,t\,ha^{-1}$ in present study) stimulated CH_4 emission from N-fertilized rice paddy soil. The largest fraction of the organic components in the WV is acids in present work and Choi et al. (2012), which might increase soil soluble C source for CH_4 generation and emission (Cai et al., 2007). According to Win et al. (2009), WV addition suppressed NH_3 volatilization from a paddy soil by preventing the transition of NH_4^+ into liquid NH_3 concentration (Sommer et al., 2003). In addition, in our study, Urea + WV10 treatment effectively suppressed N_2O emission indicating the probably lower nitrification-denitrification rate and thereby higher NH_4^+ existed in soil. Perhaps, competition of NH_4^+ for the oxidation with CH_4 by methanotrophs (Mosier et al., 1991), adversely stimulate CH_4 emission from N-fertilized rice paddy fields receiving higher load of WV. Therefore, in flooded rice systems, the interactions between WV application and the CH_4 cycle are complex with different processes when WV was applied at different levels, which needs further research in the future.

3.3. Total global warming potential (GWP_t) of N_2O and CH_4

Data of GWP_t of N_2O and CH_4 from rice paddy soil were organized in Fig. 5. GWP_t of N-fertilized paddy soil under a single rice cycle ranged from $5.24 \pm 0.53\,t\,CO_2\text{-e}\,ha^{-1}$ to $12.0 \pm 0.71\,t\,CO_2\text{-e}\,ha^{-1}$ and CH_4 contributed 61.1–84.1% of GWP_t. Interestingly, application of WV at a lower rate ($5\,t\,ha^{-1}$) mitigated GWP_t by 31.5%, as result of its positive effect on reducing both N_2O and CH_4 emissions (Figs. 2 and 4). However, if applied with larger amount ($10\,t\,ha^{-1}$), WV reduced N_2O

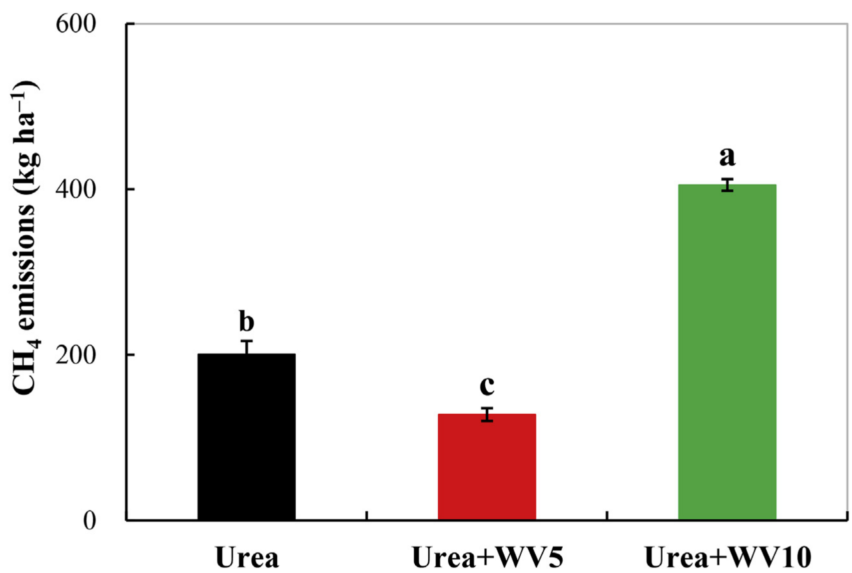

Fig. 4. Cumulative CH_4 emissions from paddy soil over a single rice cycle as impacted by wood vinegar (WV) addition. The *bars* represent the standard deviation of triplicates ($n = 3$). Different *Lowercase letters* indicate significant ($P < 0.05$) differences among treatment means.

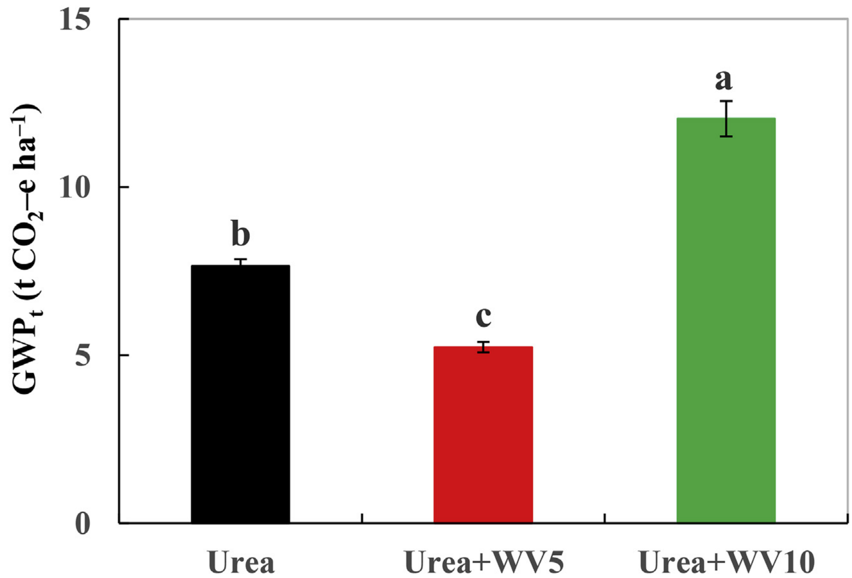

Fig. 5. Total global warming potential (GWP_t, CO_2-equivalents) of N_2O and CH_4 from paddy soil over a single rice cycle as impacted by wood vinegar (WV) addition. The *bars* represent the standard deviation of triplicates (n = 3). Different *Lowercase letters* indicate significant (P < 0.05) differences among treatment means.

but increased CH_4 emission at a much larger scale (Figs. 2 and 4). Simultaneously considering the fact that CH_4 contributed 61.1–84.1% of GWP_t in N-fertilized rice paddy soil, WV adversely increased GWP_t by 57.2% (Fig. 5). Therefore, 5 t ha^{-1} WV is suggested for incorporating into rice paddy soil because it can suppress both N_2O and CH_4 emissions and thereby the GWP_t.

Present work evidenced that WV applications with different rates has contrast impacts on GWP_t from rice paddy soil received N fertilizer, which was mainly as results of its contrary impact on CH_4 emission. In rice paddy soil, WV could change the C and N fates by influencing soil pH (Win et al., 2009), soil microbial community structure and enzyme activity (Lu et al., 2015) and the crop growth (Lashari et al., 2013; Polthanee et al., 2015). It is easily understood that WV application at varied rates has diverse impacts on these aspects related to the eventual CH_4 and N_2O emissions. Therefore, the optimal rate of soil amendment (such as MV in the present study) should be firstly investigated before its large-scale application.

4. Conclusion

Soil column experiment was conducted to evaluate the impact of WV application on N_2O, CH_4 emissions and the GWP_t from N-fertilized rice paddy soil. Both N_2O and CH_4 emission flux patterns were not changed after WV incorporation. However, WV application indeed influenced GHGs emission rates, as well as the cumulative emission loads. When applied at lower rate (5 t ha^{-1}), WV simultaneously suppressed N_2O and CH_4 emission, and the corresponding GWP_t. Reduced N_2O, but dramatically increased CH_4 emissions were observed in rice paddy soil receiving 10 t ha^{-1} WV, leading to a higher GWP_t. Therefore, application of WV at an optimal rate was recommended for its function of both reducing N_2O and CH_4 emissions from N-fertilized rice paddy soil. However, its impact mechanisms and long-term effects need further investigations.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.05.015.

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