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Effects of fertilizer regimes on greenhouse gas emissions in a Gray Luvisol from central Alberta, Canada

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Abstract. This study investigated how different fertilizer regimes with a long-term repeated N application experiment affected on the soil Greenhouse gases (GHGs) emissions in a Gray Luvisolic from central Alberta, Canada. The results showed fertilizer regimes significantly influenced on GHGs emissions. The higher emissions of N_2O and CH_4 induced by NPKS soils were due to the superior utilization rate of mineral N and organic N. The CO_2 emission rates indicated that soil organic carbon (SOC) was upper after Manure application than that after NPKS and Lime applications. The lowest Global Warming Potential (GWP) and highest pH of Lime soils suggested that liming application cause a significant decrease of GHGs by changing soil properties, such as pH. As for CH_4 emissions during the incubation period, NPKS soil acted as sources, whereas the Lime and Manure soils acted as CH_4 sinks. In addition, the fertilization history with higher SOC stocks in the Manure soils did not affect higher N_2O emissions. In conclusion, this research showed liming application could be a better policy for improving soil properties in the acid Gray Luvisol from central Alberta, but liming may result in lower emissions of N_2O and CO_2 than the treatment of mineral fertilizer or farmyard manure.

1. Introduction

It is known that greenhouse gases (GHGs) includes carbon dioxide (CO_2), nitrous oxide (N_2O) and methane (CH_4), which are the important contributors to the global warming. Although the increasing rates of N_2O and CH_4 contents in the air are significantly slower than those of CO_2, their values of the global warming potential (GWP) over a 100-year time scale are 28 and 265 times higher than that of CO_2, respectively [1]. Agricultural soils are regarded as the primary resources of anthropogenic GHGs [2-4]. Soil management practices in agricultural systems, such as fertilization, may influence the release of soil carbon (C) and nitrogen (N) to the atmosphere by changing soil pH and microbial activities that in turn affect C and N cycle [5,6].

Many research indicated fertilizer regimes have significant effects on GHGs emissions in agricultural soils. Numerous studies indicated that N_2O emissions from inorganic fertilizers (such as NPK or urea) were significantly higher than those from organic manures in different cultivated soils [7,8]. However, other reports suggested that organic manures improve N_2O emissions in comparison with with inorganic manure applications [3,9,10]. In addition, there was no remarkable influence on the emissions of N_2O between inorganic fertilizer and organic manure [11]. Meanwhile, Barton *et al*

[12,13] and Cheng *et al* [14] showed that lime applications could lower GHGs emissions by decreasing N_2O fluxes and increasing CH_4 uptake. In spite of many research have analyzed the effects on soil GHGs emissions from different fertilizer regimes between organic and inorganic manures, it is very few reports about the fertilization experiments over decades [6,15]; thus it is important to study soil GHGs emissions in response to long-term fertilizer applications [6,10,16].

The gray-wooded soils occur mainly in the northern interior plains of Manitoba, Saskatchewan and Alberta in Canada, which are now known as Gray Luvisolic soils. The Breton Classical Plots of a Gray Luvisolic soil, which were established in central Alberta in 1930, provide a model of how diverse crop and fertilizer practices affect typical Gray Luvisolic soils. And 8 fertility treatments were founded in the Plots since 1980, including Control, Manure, NPK, Lime, NSK, PKS, NPS and NPKS (NPK in combination S-fertilizer) [17]. In addition, the chronic S-fertilization enhanced carbon fixation and reduces N_2O emissions in the Breton Classical Plots [18]. Meanwhile, in agricultural research, laboratory incubation of repacked soils is an important tool for elucidating GHGs emissions from agroecosystems [19]. Therefore, this paper is aimed to estimate the impacts of the standing fertilizer treatments (i.e. Manure, Lime and NPKS as compared with Control) on short-term (30 days) GHGs emission from Gray Luvisolic soils in the Breton classical plots by laboratory incubation experiments.

2. Material and methods

2.1. Soil sampling

Soil samples were collected from the Breton classical plots of the long-term fertilization test. The plots located in central Alberta, which is 53°07′N, 114°28′W and its elevation is 830 m. The soils are classified as a Gray Luvisol, which is typic Cryoboralf named by United States Department of Agriculture taxonomy. The silty loam soil locates in the Breton Plots, and it has a particle size distribution, which is 120 g clay kg^{-1}, 620 g silt kg^{-1} and 260 g sand kg^{-1} [20]. And in the plots, there are 8 fertilizer treatments since 1980 [17].

These field specimens were gathered from four fertilizer treatments (i.e. the Control, Manure, NPKS and Lime) in October, 2015 (table 1). In each treatment, a composite soil sample (about 2000 g) was gathered using soil augers (3.5-cm diameter) in the 0-10 cm depth from 4 random points within each point [21]. With a 2 mm sieve, fresh soil samples were passed through, then removed coarse fragments and roots, and manually homogenized in the laboratory. Then soil samples were stored at 4 °C until needed for the cultivating experiment. The main properties of the four soil samples are given in table 2.

Table 1. The fertilizer treatments in the Breton classical plots in this study.

Treatment	N (kg/ha)	P (kg/ha)	K (kg/ha)	S (kg/ha)
Control	0	0	0	0
Manure	#			
NPKS	*	22	46	5.5
Lime	0	0	0	0

N application via manure depends on the rotation (wheat-fallow: 90 kg N ha^{-1} during cropped years).
*N amounts depend on the crop and its place in the rotation (wheat on fallow: 90 kg N ha^{-1}).

Table 2. Chemical and physical properties of the soil studied (mean±SD).

Soil samples	pH	TC (g kg^{-1})	TN (g kg^{-1})	C/N	WSOC (mg kg^{-1})	WSON (mg kg^{-1})
Control soils	6.28±0.11	5.36±0.24	0.48±0.08	11.17±0.42	41.58±2.85	16.08±3.03
Manure soils	6.27±0.13	10.81±0.46	0.82±0.13	13.18±0.58	93.11±8.85	28.25±3.80
NPKS soils	4.70±0.06	5.28±0.16	0.43±0.09	12.28±0.61	41.92±4.18	47.08±9.43
Lime soils	6.85±0.05	6.29±0.15	0.45±0.06	13.98±0.86	45.91±3.63	40.97±8.36

2.2. Soil incubation and gas sampling

For each fertilizer treatment, 16 soil samples, of which each sample is 25.0 g (dry-weight basis), were placed in conical flasks with 250-mL volume, respectively. Then, with a mini-pipette, each flask was added into deionized water evenly over the soil surface, and makes the soil 60% water-holding capacity (WHC). With rubber stoppers, these flasks were sealed and incubated 30 days at 20°C in the dark. To keep the aerobic condition in the flasks, these flasks were aerated for 5 min each day during the incubation period. And by using a mini-pipette and deionized water was added to compensate for waster loss in the flasks every 3 days.

On days 1, 3, 6, 9, 12, 15, 18, 21, 24, 27 and 30, 4 flasks were taken randomly as replicates in each treatment. Using a gas-tight syringe, after the flasks had been sealed with rubber stoppers for 0 and 24 hours, a 20-mL gas sample was gathered from each flask. Then the gas sample was transferred to an evacuated gas-tight vial (12.5 mL) for GHGs analysis by a Varian CP-3800 gas chromatograph (Varian Canada, Mississaugua, Canada) with an electron capture detector. The detailed configuration and working condition of the gas chromatograph was described by Paterson et al. (2004). In addition, for each series and treatment, a set of 15 additional flasks were prepared in triplicate and stored in the same conditions On days 0, 6, 14, 22 and 30 in the incubation period, triplicate flasks from each treatment were randomly selected to test soil NH_4^+-N and NO_3^--N, water-soluble organic C (WSOC) and N (WSON).

2.3. Soil properties analysis

Soil samples were shaken for 20 min with deionized water, which was 1:5 (mass: volume ratio). After the mixture stand for 5 min, the pH of the samples was measured using a pH meter (DMP-2 mV, Thermo Orion, USA). The concentrations of total C and N were analyzed by a CN analyzer (NA Series 2, CE Instruments, Italy). By 2 mol L^{-1} KCl solution at a ratio of 1:5 (w:v), soil samples were extracted to analyze soil mineral N, including NH_4^+-N and NO_3^--N, by the method of Li *et al* [22] and Miranda *et al* [23]. During the incubation (i.e., on days 0, 6, 14, 22 and 30), WSOC and WSON were conducted by extracting 10 g (dry-weight basis) of soil with 50 mL of deionized water, and centrifuged for 20 min at 4000×g. Then each sample was filtered through a 0.45-μm membrane filter. Lastly, SOC and SON concentrations were analyzed by a TOC-V Total Organic Carbon Analuzer (Shimadzu Crop, Kyoto, Japan).

2.4. Calculation and statistical analysis

Emission rates (fluxes) of N_2O, CO_2, and CH_4 were calculated by the following equation (1):

$$F = \frac{\rho \times \Delta c \times V \times 273}{W \times \Delta t \times (273 + T)} \quad (1)$$

where F is the flux of N_2O (ng N_2O-N kg^{-1} h^{-1}), CO_2 (mg CO_2-C kg^{-1} h^{-1}), and CH_4 (ng CH_4-C kg^{-1} h^{-1}); ρ is the density of N_2O, CO_2, or CH_4 under standard state; Δc is the change of gas concentration between incubation time of 0 and 24 h (ppbv h^{-1} or ppmv h^{-1}); V is the head space volume of the conical flasks (mL); W is the dry weight of soil (kg); Δt is the interval between two measurements; and T is the incubation temperature (°C) [24].

Provided a constant flux rate of each gas sampling from the beginning until the next gas sampling, the cumulative N_2O, CO_2 and CH_4 emissions in each replication were calculated from the integrated daily fluxes. The cumulative emissions of N_2O, CH_4 and CO_2 were calculated as the following equation (2) [25]:

$$\text{Cumulative } N_2O \left(\text{or } CH_4,\ CO_2\right) \text{ emission} = \sum_{i=1}^{n} \frac{(F_i + F_{i+1}) \times 24}{2 \times (t_{i+1} - t_i)} \quad (2)$$

where F is the N_2O or CH_4, CO_2 emission rate (flux), i is the ith measurement, the term of (t_{i+1} - t_i) is the days between two adjacent days of the measurements, and n is the total times of the measurements.

The GWPs were expressed by CO_2 equivalent flux values. And they were calculated by multiplying CH_4 emissions rates by 28 and N_2O emission rates by 265, and adding the products to the CO_2 emission rates [1].

By the SPSS 16.0 software package for Windows (SPSS Inc., Chicago, USA), all data were statistically analyzed. The effects of soil properties on each cumulative of the GHGs (N_2O, CO_2 and CH_4) emission was evaluated by one-way ANOVA, followed by the least significant difference test at $P<0.05$. All figures are made by Origin 9.0 (Origin Lab Inc, Corporation, USA).

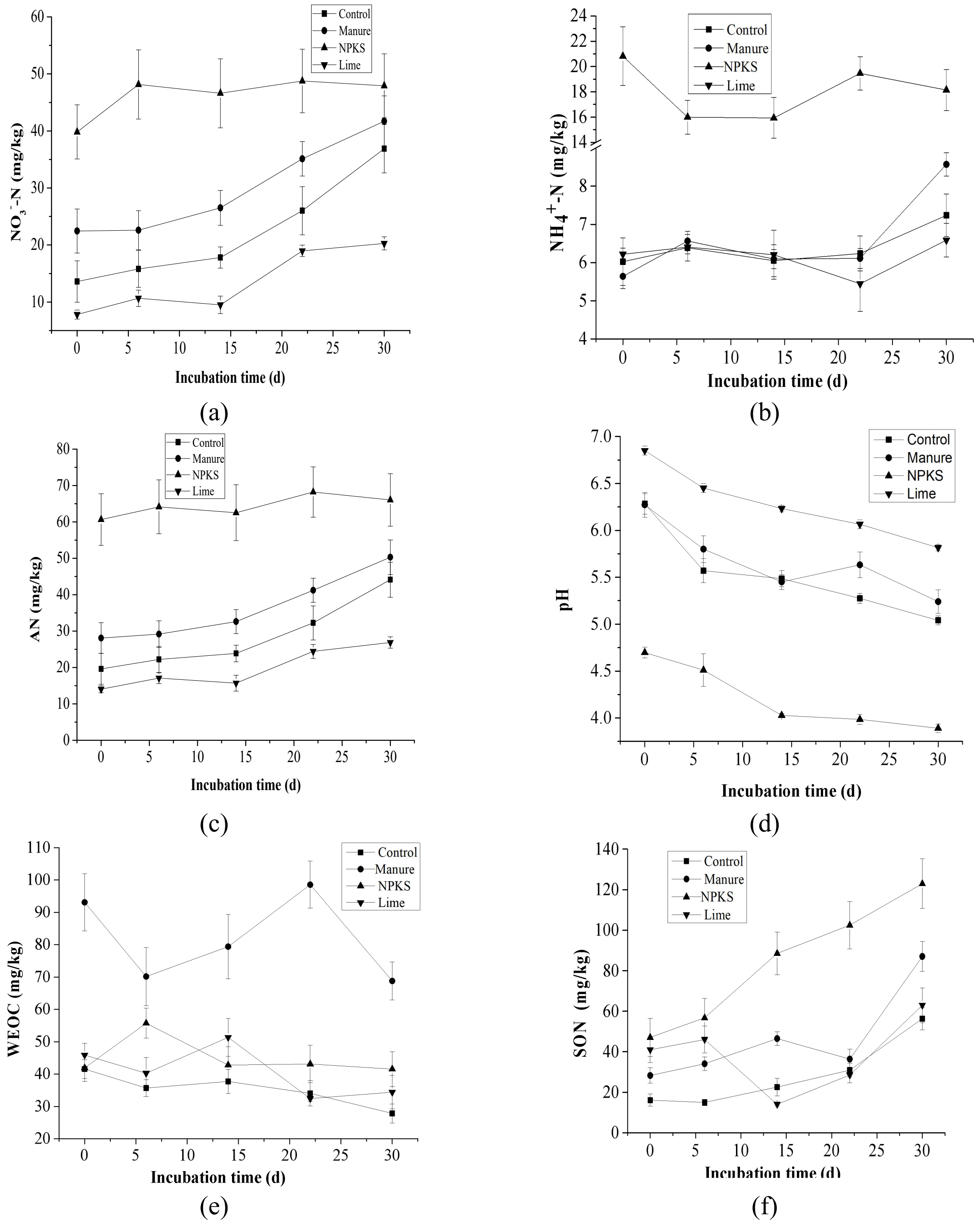


Figure 1. Temporal variation of NH_4^+-N, NO_3^--N, available nitrogen (NH_4^+-N+NO_3^--N), pH, SOC and SON contens in different treatments at 60% water holding capacity. AN: Vertical bars indicate standard error of the means (n=4).

3. Results

3.1. Soil properties

During the whole incubation, the means NO_3^--N concentrations of NPKS, Manure, Control and Lime soils were 46.2, 29.7, 22.0 and 13.4 mg kg^{-1}, respectively (figure 1(a)). Meanwhile, the means of NH_4^+-N concentrations of the four treatments were 18.1, 6.6, 6.4 and 6.2 mg kg^{-1}, respectively (figure 1(b)). These indicated that NO_3^--N was the dominant mineral nitrogen (N). In all treatments, NO_3^--N concentrations increased significantly throughout the incubation period (figure 1(a)). Across the incubation period, soil mineral N in Control, Manure and Lime soils were significantly lower than those in NPKS soils (figure 1(a)-1(c), P<0.05). And soil mineral N had significant positive effects on N_2O and CH_4 cumulative emissions, but not CO_2 (P< 0.05; table 3). The pH of Lime, Manure, Control and NPKS soils were 6.3, 5.7, 5.5 and 4.2, respectively (figure 1(d)), indicating that Lime increased significantly pH. And pH had remarkable negative impacts on N2O cumulative emissions (table 3).

During the whole incubation, the means WSOC concentrations in the Manure, NPKS, Lime and Control treatments were 82.0, 45.0, 40.9 and 35.4 mg kg^{-1}, respectively (figure 1(e)), indicating that the Manure soils had significantly greater WSOC compared with other treatments. For one thing, the means of SON were 83.6, 46.4, 38.5 and 28.2 mg kg^{-1} in NPKS, Manure, Lime and Control treatments respectively (figure 1(f)), indicating that the NPKS soils had significantly higher SON compared with other soils.

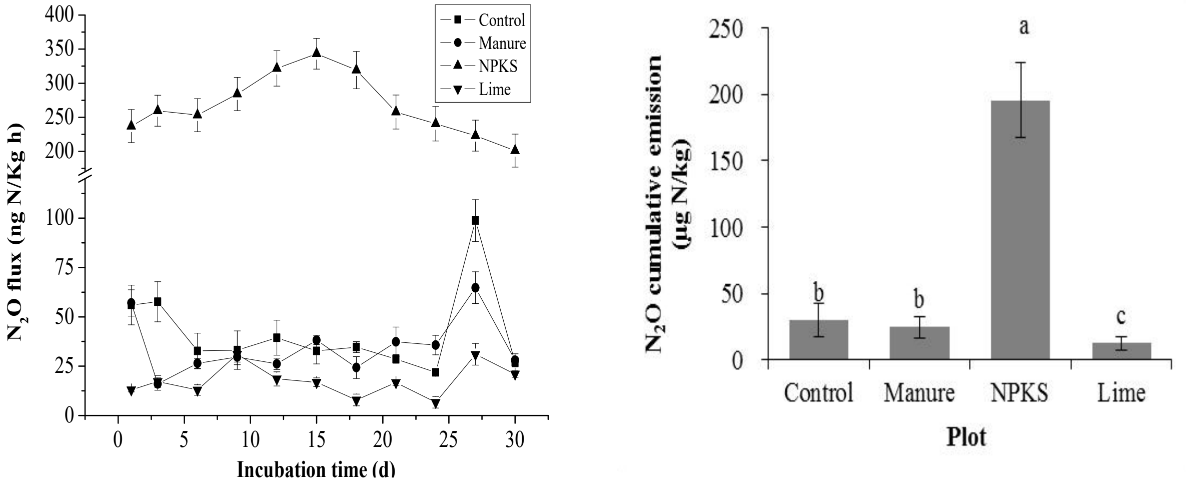
Table 3. Correlations between cumulative emissions of N_2O, CO_2, CH_4 cumulative emissions and soil properties (mean values) during a 30-d incubation.

Gas	pH	NO_3^--N (mg kg^{-1})	NH_4^+-N (mg kg^{-1})	AN (mg kg^{-1})	WSOC (mg kg^{-1})	WSON (mg kg^{-1})
N_2O cumulative emission (μg N kg^{-1})	-0.897**	0.804**	0.974**	0.880**	-0.131	0.718**
CO_2 cumulative emission (mg N kg^{-1})	0.158	0.011	-0.279	-0.068	0.839**	-0.328
CH_4 cumulative emission (μg C kg^{-1})	-0.622	0.578*	0.840**	0.671**	-0.307	0.709**

* Correlation is significant at the 0.05 level; ** Correlation is significant at the 0.01 level.

3.2. N_2O emissions

For the NPKS soils, from the first day to the fifteenth day, the temporal pattern of the N_2O emission increased gradually and then declined little by little to the end of the 30-day incubation, However, the N_2O fluxes in Control, Manure and Lime soils remained relatively constant during the whole incubation except on day 27 (figure 2(a)). During the whole incubation, the N_2O emission rates ranged from 201.0 to 343.3 ng N kg^{-1} h^{-1} in the NPKS soil, while Control, Manure and Lime soils were from 21.8 to 98.7, 16.0 to 64.8 and 6.7 to 31.0 ng N kg^{-1}, respectively. In addition, the NPKS soils had significantly higher N_2O cumulative emissions than Control, Manure and Lime soils (figure 2(b)). N_2O cumulative emissions were not significant different (30.3 and 25.0 μg N kg^{-1}, respectively) between the control and Manure treatments, while the values was significantly higher than that of Lime treatment (12.6 μg N kg^{-1}).

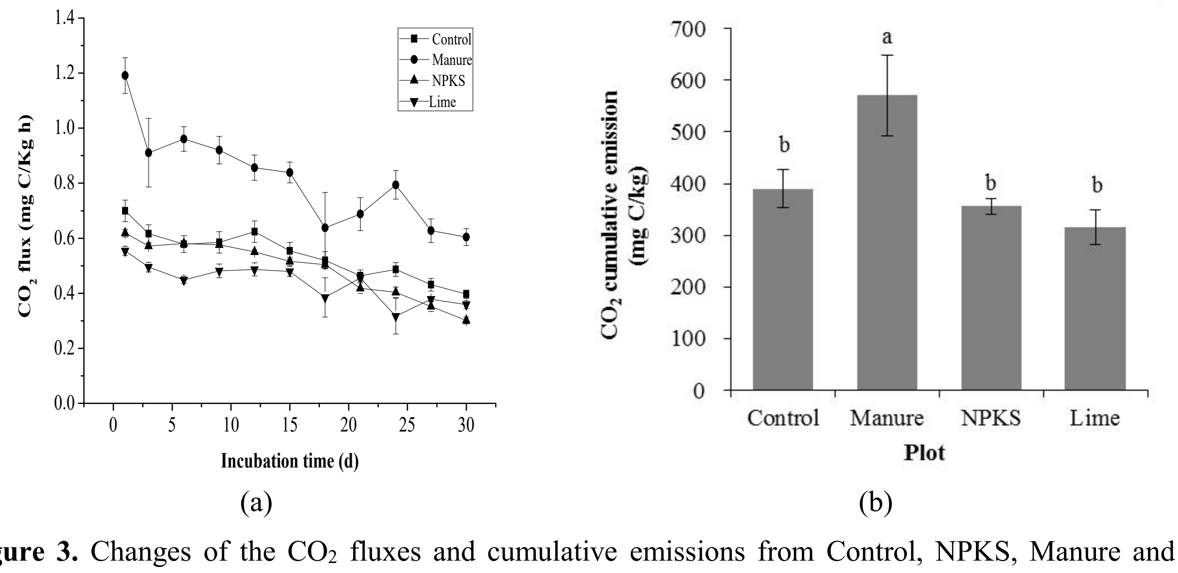


(a) (b)

Figure 2. Changes of the N_2O fluxes and cumulative emissions from Control, NPKS, Manure and Lime soils at 60% water holding capacity. Vertical bars indicate standard error of the means (n=4).

3.3. CO_2 emissions

As for the CO_2 fluxes, it is a general trend that the values in the first day arrived highest in the four treatments and those subsequently decreased until the incubation come to an end (figure 3(a)). This indicated the available carbon for mineralization declined step by step with time. Fertilizer treatments affected significantly on the CO_2 fluxes, and CO_2 fluxes from Manure soils exceeded outstandingly to Control, NPKS and Lime soils. The mean values of the CO_2 emissions for the Manure, Control, NPKS and Lime soils were 0.82, 0.54, 0.49 and 0.44 mg C kg^{-1}, respectively (figure 3(a)). In addition, the Manure soils had significantly higher CO_2 cumulative emissions than the other soils, but there were no significant differences among the control, NPKS and Lime treatments (393.3, 355.7 and 316.3 mg C kg^{-1}, respectively, figure 3(b)).



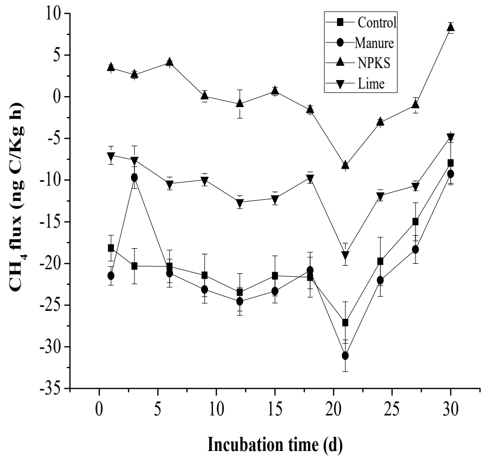
(a) (b)

Figure 3. Changes of the CO_2 fluxes and cumulative emissions from Control, NPKS, Manure and Lime soils at 60% water holding capacity. Vertical bars indicate standard error of the means (n=4).

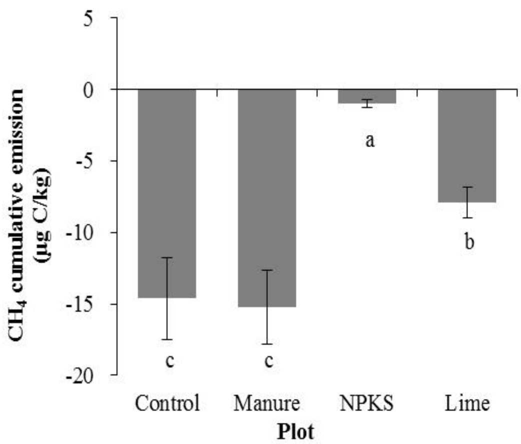
3.4. CH_4 emissions

In general, the CH_4 fluxes firstly decreased gradually from the first day to the 21st day and then increased gradually during the remainder of the incubation (figure 4(a)). The mean values of the CH_4

emissions for the NPKS, Lime, Control and Manure soils were 0.4, -10.5, -19.7 and -20.5 ng C kg^{-1}, respectively (figure 4(a)), indicating the NPKS soil acted as sink for atmospheric CH_4 during the incubation period, while the Lime, Control and Manure soils acted as sinks for atmospheric CH_4. In addition, the NPKS soils had significantly higher CH_4 cumulative emissions than the other soils (figure 4(b)). CH_4 cumulative emissions were not significant different (-14.6 and -15.3 ng C kg^{-1}, respectively) between the control and Manure treatments, but was significantly lower than that from Lime treatment (-7.9 ng C kg^{-1}).



(a)



(b)

Figure 4. Changes of the CH_4 fluxes and cumulative emissions from Control, NPKS, Manure and Lime soils at 60% water holding capacity. Vertical bars indicate standard error of the means (n=4).

3.5. Global warming potential

The contribution to GWP of the three GHGs decreased in the following order by CO_2, N_2O and CH_4 from high to low in all treatments (figures 2-4). The highest value of GWP (0.58 g CO_2 equivalent/kg) occurred in the Manure soils, the lowest value in the Lime soils (0.32 g CO_2 equivalent/kg), and the intermediate value in the Control and NPKS soils (0.40 and 0.41 g CO_2 equivalent/kg, respectively. figure 5).

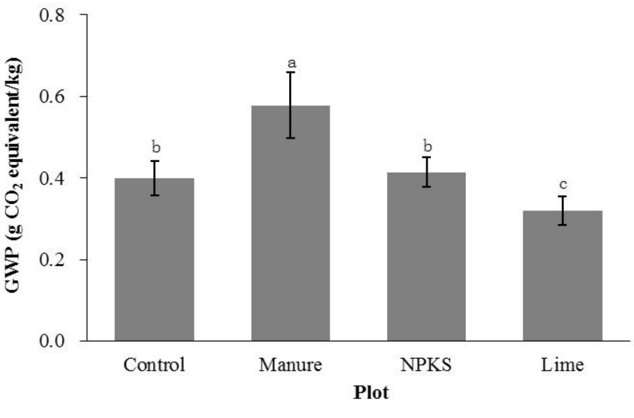


Figure 5. Total GWPs of N_2O, CH_4 and CO_2 during a 30-day incubation. Different letters in the different treatments represent significant difference at $P<0.05$. Vertical bars indicate standard error of the means (n=4).

4. Discussion

Previous research reported that fertilizer regimes in agricultural system had significant effects on N_2O emissions in soils [26,27]. In this study, fertilizer treatments affected the cumulative N_2O emissions, which decreased in the following order by NPKS, Control, Manure and Lime (figure 2), in line with these studies of Ding *et al* [7], Zhang *et al* [6] and Louro *et al* [28]. Mørkved *et al* (2007) reported that the highest soil N_2O fluxes from the NPK with straw treatments had a lower pH than those from manure applications. Similarity, this study showed that N_2O emissions had a negative and significant correlation with soil pH. And the NPKS treatment had the highest N_2O emissions with lowest soil pH compared with other three treatments. In contrast, N_2O emissions of NPKS treatments were higher than those from Manure in this study. This result was according with other studies in sandy soils, which indicated that N_2O emissions of inorganic fertilizer treatments were higher than those of Manure [8,29,30].

N_2O emissions in the soils mainly come from nitrification denitrification under anaerobic conditions and under aerobic conditions [31,32]. Meanwhile, under moderately moist conditions, the predominant N_2O-producing process (aerobic, WHC 60%) is generally reduced nitrification [33], which just likes the incubation condition in this study. In addition, the NPKS treatment had the highest N_2O emissions with lowest soil pH compared with other three treatments, which was studied by Kitzler *et al* [34] and Brumme and Beese [35]. This is likely because that liming application increased pH (figure 1(d)) and might provide an adverse condition for nitrification in the acid Gray Luvisol in central Alberta. Therefore, one of the most important factors to decrease N_2O emissions was likely from the increasing of soil pH by liming in the Gray Luvisolic soils [24].

Many researches showed that there were significant correlations between mineral N and N_2O emissions in soils [3,36-38]. As the mineral N is the primary substrate for N_2O production in soils, an increase in mineral N is generally associated with higher N_2O emissions [39]. In the present study, both NH_4^+-N and NO_3^-- N concentrations were significantly and positively correlated with N_2O emissions (table 3), suggesting that that the high concentration of available N in the NPKS soils was responsible for the dramatic increase in N_2O fluxes in the soils [24,39,40]. In addition, this study showed that Manure soils had significantly greater SOC but lower N_2O emissions compared with NPKS soils, indicating that the increasing SOC of Manure applications may be an effective way to cut down N_2O emissions in the Gray Luvisolic soils.

CO_2 emission in the soils was caused by microbial respiration in the laboratory incubation, which was mainly influenced by the discharge of readily decomposable SOC [33,41,42]. This study showed that CO_2 cumulative emissions were significantly, positively affected by SOC (table 3). Moreover, we found that fertilizer treatments had a highly significant effect on the CO_2 fluxes emissions, that CO_2 fluxes and SOC in the Manure soil significantly greater than those in the Control, NPKS and Lime soils throughout the incubation period. This conclusion was reported by previous studies [3,6,43]. In contrast, the Lime soil had the lowest CO_2 emissions and the lowest GWP indicated that Lime application may be the most effective measure in all treatments to mitigate CO_2 emissions from Gray Luvisolic soils in central Alberta. In addition, on the first day, CO_2 fluxes in all treatments attained the highest values and then subsequently decreased in the end of the incubation period (figure 3(a)). This was due to higher microbial activity and the rate of soil respiration at the beginning of the incubation [44,45].

Soils CH_4 emissions originate from the process of microbial decomposition under strictly anaerobic conditions [46,47]. During the incubation period, NPKS soils acted as sources for atmospheric CH_4, while the Lime, Control and Manure soils might be sinks in this study (figure 4). However, anaerobic microsites in NPKS soils could induce weak fluxes of CH_4 at 60% WHC. Meanwhile, CH_4 emissions are influenced by soil properties, such as soil pH, NH_4^+-N and SOC [48-50]. In addition, CH_4 emissions have a direct relationship with soil mineral N by preventing the oxidation of CH_4 to the atmosphere [51]. In this study, soil mineral N had significant positive effects on N_2O and CH_4 cumulative emissions (table 3). This result was not in agreement with other reports, which were negative relationships of CH_4 uptake and soil NH_4^+-N concentration [52,53]. This phenomenon may due to the higher amount of mineral N, which induced the positive emissions from the Gray Luvisolic soils in central Alberta.

In this study, the contribution of the three GHGs to GWPs increased in the following order by CH_4, N_2O and CO_2 (figures 2-4), which indicated that the greenhouse effect for the releases of CO_2, N_2O and CH_4 mainly depended on CO_2 emissions from the Gray Luvisolic soils to atmosphere [54-56]. Meanwhile, the GWPs of the three GHGs were significantly influenced by fertilizer treatments, and the highest GWPs were observed for the Manure soils (figure 5).

5. Conclusion

In this study, fertilizer regimes had significant effects on soil GHGs emissions, which higher N_2O and CH_4 emissions were induced by NPKS application and higher CO_2 emissions by the Manure applications. Meanwhile, the lowest GWP and GHGs emissions of Lime soils suggested that it may be a preferred fertilizer strategy for mitigating soil greenhouse effect from the application of liming in the acid Gray Luvisolic soils in central Alberta. In addition, higher N_2O and CH_4 emissions from NPKS soils were due to a high availability of mineral N and organic N. However, further studies in the field environments should be carried out to confirm the result from the laboratory incubation experiments.

Acknowledgments

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