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Enhancement of farmland greenhouse gas emissions from leakage of stored CO_2: Simulation of leaked CO_2 from CCS

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HIGHLIGHTS

- Relationship between CO_2 leakage and CH_4 and N_2O emissions was examined.
- Geologically stored CO_2 leaking into surface soil enhances CH_4 and N_2O emissions.
- GWP of additional CH_4 and N_2O is negligible compared with amount of leaked CO_2.
- Significant increase of CH_4 and N_2O emissions from soil could indicate CCS leakage.

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ABSTRACT

The effects of leaked CO_2 on plant and soil constitute a key objective of carbon capture and storage (CCS) safety. The effects of leaked CO_2 on trace soil gas (e.g., methane (CH_4) and nitrous oxide (N_2O) emissions in farmlands are not well-understood. This study simulated the effects of elevated soil CO_2 on CH_4 and N_2O through pot experiments. The results revealed that significant increases of CH_4 and N_2O emissions were induced by the simulated CO_2 leakages; the emission rates of CH_4 and N_2O were substantial, reaching about 222 and 48 times than that of the control, respectively. The absolute global warming potentials (GWPs) of the additional CH_4 and N_2O are considerable, but the cumulative GWPs of the additional CH_4 and N_2O only accounted for 0.03% and 0.06%, respectively, of the cumulative amount of leaked CO_2 under high leakage conditions. The results demonstrate that leakage from CCS projects may lead to additional greenhouse gas emissions from soil; however, in general, the amount of additional CH_4 and N_2O emissions is negligible when compared with the amount of leaked CO_2.

1. Introduction

Carbon capture and storage (CCS) technology may be strategically important for mitigating global greenhouse gas (GHG) emissions (IPCC, 2005). CCS is particularly important for coal-based GHG emitters, such as China (Sun, 2006; Li et al., 2011). Preliminary estimates show that the CO_2 storage capacity in China is in the hundreds of billions of tons. Moreover, more than 12 demonstration projects have been established over the past few years (Zhang et al., 2013). Naturally, one of the key concerns associated with CCS is CO_2 escaping from the deep geological storage formation (Lions et al., 2014) and adversely impacting the environment (Damen et al., 2006; Beaubien et al., 2008; Noble et al., 2012).

Agriculture accounts for 52% and 84% of global anthropogenic CH_4 and N_2O emissions, respectively. CH_4 is produced when organic materials decompose in oxygen-deprived conditions and N_2O is generated by the microbial transformation of nitrogen in soil (Smith et al., 2008). Traditionally, CH_4 and N_2O emissions are linked to cropland management and cultivated organic soils. CO_2 leakage is also thought to have an effect on CH_4 and N_2O emissions. Leaked CO_2 eventually enters the atmosphere through the soil, leading to locally high soil CO_2 concentrations (Zhou et al., 2012). However, relatively little is known on the effects of leaked CO_2 on CH_4 and N_2O emissions in farmlands. There are no signs of CO_2 and CH_4 leakage reported from the continuous monitoring of CCS projects (Beaubien et al., 2013). Nevertheless, several studies have focused on the effects of elevated soil CO_2 on CH_4 and

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N_2O emissions. Krüger et al. (2011) collected CH_4 measurements at a vegetated pasture field in the Laacher See volcanic center in Germany. CH_4 production rates of 0.33 ± 0.007 μmol CH_4 g_{dw}^{-1} day^{-1} were reported at the vent center, with a soil CO_2 flux of more than 550 g^{-2} $days^{-1}$. These rates were much higher than those found in the control site samples with values of 0.12 ± 0.002 μmol CH_4 g_{dw}^{-1} day^{-1}, where the soil CO_2 flux dropped to about 50 g^{-2} $days^{-1}$. Ziogou et al. (2013) found CH_4 production rates of 0.016 μmol CH_4 g_{dw}^{-1} day^{-1} at the CO_2 vent (100% CO_2), which was three times higher than those at a medium site with 10% CO_2. Additionally, no CH_4 production could be detected at the reference site (<0.9% CO_2) of a natural CO_2 vent in a pasture in the Florina Basin in Greece. Furthermore, the effects of CO_2 flux on soil CH_4 were spatially restricted. Šibanc et al. (2014) showed a shift towards methanomicrobia dominated communities under elevated soil CO_2 concentrations and recorded traces of CH_4 in soil with high CO_2 concentrations. In contrast, at the Latera geochemical field in central Italy, Beaubien et al. (2008) found that the CH_4 production rate at the background site was 2.34 ± 0.35 nmol CH_4 g_{dw} $soil^{-1}$ day^{-1}, which was three times higher than the vent core (0.79 ± 0.13 nmol; high CO_2 and H_2S) and eight times higher than the transition zone (0.29 ± 0.03 nmol; high CO_2 but no H_2S). Evidently, elevated soil CO_2 is a vital factor in CH_4 and N_2O production. However, it is difficult to understand and compare these trends, owing to the contrasting results from natural analogies and the considerable environmental difference among the test sites.

In this paper, the effects of elevated soil CO_2 on CH_4 and N_2O were examined through pot experiments, which simulated the CO_2 fluxes from common to extreme leakage conditions. The specific objective was to test the hypothesis that leaked CO_2 induces additional CH_4 and N_2O emissions in farmlands and contributes to global warming.

2. Data and experimental design

2.1. Location and experimental timeline

The experiment was performed at the Experimental Station of the Chinese Academy of Agricultural Sciences, located in Beijing (39° 57′ 30″ N, 116° 19′ 43″ E). The average annual conditions are temperatures of 11–12 °C and rainfall of 640 mm. The annual sunshine hours range from 2000 to 2800 h with a frost-free period of 190–195 days and wind speed of 1.8–3.0 $m{\cdot}s^{-1}$. The region has a temperate, semi-humid monsoon climate (Wu et al., 2012). The experiment was conducted from Aug. 12th to Oct. 25th 2010 (75 days). CO_2 was injected continuously from September 12th 2010 to the end of the experimental period, in order to reproduce a constant leak (Fig. 1).

2.2. Pot experiments for simulated CO_2 leakage

The pot experiments were designed using accumulation-chamber measurements (Wu et al., 2012) to simulate a simple ecosystem. They consisted of a controlled CO_2 release device; a set of CO_2, CH_4, and N_2O flux monitoring/recording instruments; and a management system. The principles and compositions of the experiments, used to assess the impact of CO_2 leakage on CH_4 and N_2O emissions, were as follows:

(i) *Planting and management*. The maize hybrid (Jingkenuo 2000) was selected for this study. Maize was seeded on Aug. 12th 2010 and seedlings were singled out at the three-leaf stage, leaving one seedling per pot. The soil type used was cinnamon soil (0–20 cm, equivalent to Ustalf in the US soil classification system) (Stoner and Baumgardner, 1981); it was composed of clay (31.6%), silt (46.1%), sand (19.3), and gravel (3.0%). The physical and chemical characteristics of the soil are listed in Table 1. All the treatments included the application of water and fertilizers in a manner similar to the common agronomic practices in Beijing (Huang et al., 2001) (Table 2).

(ii) *Controlled CO_2 release device*. The device used to manually control the release of CO_2 consisted of experimental pots (soil chamber, permeable separator, and CO_2 gas chamber), gas duct, ball valve, gas shunt, CO_2 gas source, gas meter, and drain valve (Fig. 1). The pots used were made of 10 mm thick plastic, the gas duct was made of 3 mm soft plastic pipe, and the CO_2 source gas was supplied using a steel gas bottle filled with standard CO_2 (50 kg, 15 MP, 99.5%). Drain valves used to remove excess irrigation water were made of steel. The total irrigation water applied during the experimental period was 26 L pot^{-1}, no irrigating water was drained. The soil chamber was 33 cm in height, 40 cm in diameter at the upper portion, and 36 cm in diameter at the lower portion. The CO_2 gas chamber was 17 cm in height, 36 cm in diameter at the upper opening, and 33 cm in diameter at the lower portion. A gas permeable separator was placed between the soil and gas chambers. The CO_2 injected into the gas chamber passed through the permeable separator and moved upward until it was released from the near-surface.

(iii) *CO_2 leakage treatment setting*. The CO_2 injection flux (hereafter referred to as flux) was used as a key indicator of leakage. According to the literature, the fluxes of common natural CO_2 seepages are 1000–3000 $g{\cdot}m^{-2}{\cdot}d^{-1}$ (Beaubien et al., 2008; West et al., 2009). However, the highest recorded CO_2 flux

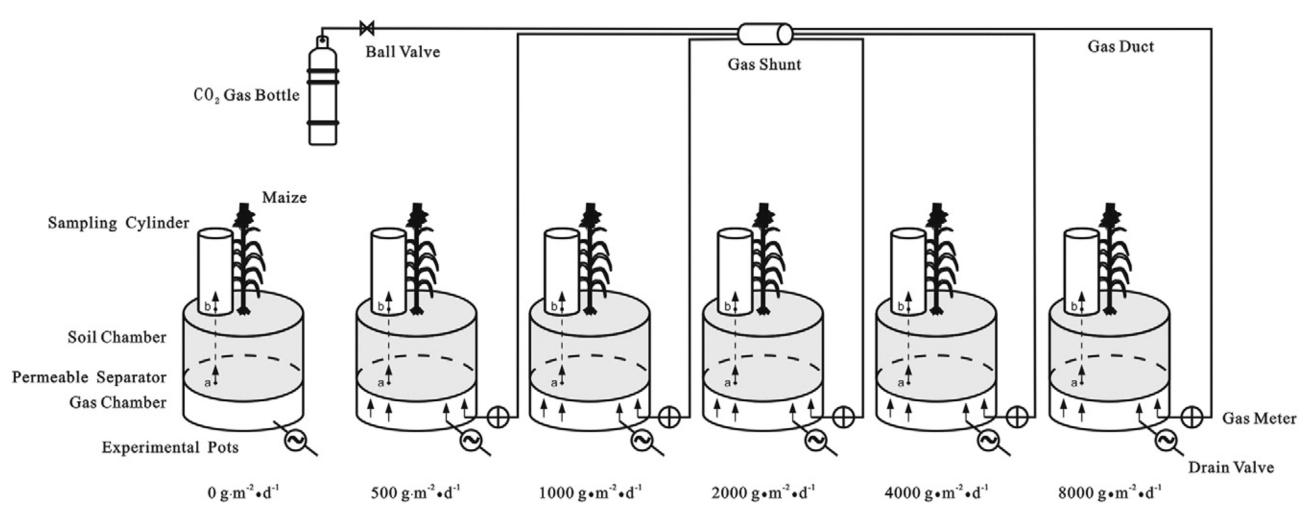

Fig. 1. Design of pot experiment for simulation of CO_2 leakage. The CO_2 injection flux was controlled at six different values; (a) experimental pots, (b) sampling cylinder.