Real-time monitoring of carbon dioxide emissions from a shallow carbon dioxide release experiment

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Abstract
This study was conducted to analyze CO₂ migration from a shallow CO₂ release experiment using a continuous soil CO₂ flux measurement system. Approximately 1.8 t CO₂ was injected from 1 to 30 June 2016 through the point sources with perforated release wells laid at 2.5-m soil depth. Using LI-8100A instruments, CO₂ concentration, CO₂ flux, soil temperature, soil moisture, relative humidity, and atmospheric pressure were continuously measured every 30 min at 0, 1.5, 3.0, 4.5, and 6.0 m from the well from 29 May to 4 August 2016. Typically sensors for soil temperature and moisture were installed at 5-cm depth, and CO₂ concentration, relative humidity, and atmospheric pressure were measured at the chambers. The CO₂ flux was not maximum directly above the release well. Carbon dioxide flux at 6.0 m from the well was similar to the background level. The relationship between CO₂ flux and environmental factors, described using a temporal correlation analysis, indicated that CO₂ flux was primarily driven by soil temperature and had the inverse correlation with relative humidity and atmospheric pressure. Heavy rainfall inhibited in-soil CO₂ migration by filling the soil pore with water. The anomalously high CO₂ flux detected at 1.5 m from the well may have been caused by the associated permeability structure, in which a permeability discrepancy leads to the vertical or horizontal flow of in-soil CO₂. These findings from this shallow CO₂ release experiment should be considered as basic information to characterize and model the in-soil CO₂ transport related to CO₂ leakage.

1 | INTRODUCTION

International concern about global warming is continually increasing because of its wide impacts on the environment. Increasing concentrations of atmospheric greenhouse gases including CO₂, CH₄, and N₂O have resulted in attention to global warming and associated climate change. Among these gases, CO₂ is considered to be the main contributor to climate change (IPCC, 2005). Therefore, C capture and storage (CCS) has been prioritized as a promising alternative that can greatly reduce CO₂ emissions. This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited. © 2020 The Authors. Vadose Zone Journal published by Wiley Periodicals, Inc. on behalf of Soil Science Society of America

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emitted from various anthropogenic sources (IEA, 2013; IPCC, 2005; Koornneef, Ramirez, Turkenburg, & Faaaij, 2012). Carbon capture and storage is a process consisting of capturing anthropogenic CO\textsubscript{2} and transporting it for safe and permanent storage in underground geologic reservoirs.

Despite the environmental benefits, there is considerable concern about potential risks of leakage, which might affect the natural environment and local human society (Haszeldine, 2009; Krüger et al., 2011). Therefore, environmental monitoring should be required to detect, quantify, and forecast potential CO\textsubscript{2} leakage (Klusman, 2011; Ko, Yun, & Chung, 2016; Lewicki, Hilley, Dobek, & Spangler, 2010) and to confirm that stored CO\textsubscript{2} does not leak out into the atmosphere (Kim et al., 2018).

Many studies have investigated the potential effects of CO\textsubscript{2} leakage on ecosystem functions by developing monitoring techniques to detect possible leakage near the soil surface. These previous studies were based on artificial CO\textsubscript{2} leakage or injection field experiments, namely the Artificial Soil Gassing and Response Detection project (ASGARD) in United Kingdom (Smith et al., 2013), the CO\textsubscript{2} Field Laboratory site in Norway (Jones et al., 2014), the Ginninderra experiment in Australia (Feitz et al., 2014), and the Zero Emissions Research and Technology project (ZERT) in the United States (Spangler et al., 2009). Such studies indicated that the diffusion of in-soil CO\textsubscript{2} gas is greatly affected by the physical conditions of the soil (Kim et al., 2017), and a CO\textsubscript{2} leakage detection system should be site specific given that each country exhibited highly different soil conditions (Kim et al., 2018). Soil properties vary even within a small area. Local variations in soil characteristics may determine the main pathway of CO\textsubscript{2} gas leakage. Thus, as it is difficult to analyze the soil characteristics throughout a CCS site, monitoring techniques are necessary for surface monitoring project (Schroder, Zhang, Zhang, & Feitz, 2016). Even though there is a range of suitable monitoring technologies, uncertainty still remains on how to identify the appropriate monitoring tasks, how to set the monitoring performance requirements, and how to decide the appropriate diversity and intensity of monitoring (Bourne, Crouch, & Smith, 2014).

The Korea-CO\textsubscript{2} Storage Environmental Management (K-COSEM) research center established the Environmental Impact Assessment Test Facility (EIT) in Eumseong, South Korea (Jun, Cheon, Yi, & Yun, 2017). The shallow CO\textsubscript{2} release experiment was conducted to measure CO\textsubscript{2} concentration and fluxes, and to analyze the migration of in-soil CO\textsubscript{2}. An in situ continuous measurement instrument, the LI-8100A (LI-COR), was used to investigate and characterize the variations of in-soil CO\textsubscript{2} and the relationship between CO\textsubscript{2} flux and environmental factors during the artificial CO\textsubscript{2} release experiment. Here, we present our investigations of the simulated CO\textsubscript{2} leakage from sources near the soil surface, in order to attain further insight into the surface expression of CO\textsubscript{2} and the suitability of several near-surface monitoring methods.

Results from the same release experiment have already been published. Kim et al. (2018) characterized the spatial distribution of CO\textsubscript{2} leakage. As the results, Kim et al. reported the (a) feature of CO\textsubscript{2} gas diffusion within a 10-m radius of surface leakage, (b) the relevance of long-term monitoring for the environmental risk assessment, and (c) the nonlinear regression and CO\textsubscript{2}/O\textsubscript{2} ratio methods to detect low-level CO\textsubscript{2} leakage. In addition, Jeong et al. (2017) developed a data-driven method to predict CO\textsubscript{2} leakages. As a result, (a) an optimal data-driven model capable of predicting the change in CO\textsubscript{2} concentration was obtained using actual field-observed datasets. (b) The developed ensemble method provides improved predictions, and the uncertainty of estimations, is appropriately quantified. (c) That method can be practically used by decision-makers operating CO\textsubscript{2} storage sites because of low computational costs. However, this study was to indicate the effects of environmental factors on CO\textsubscript{2} migration and the spatial distribution of in-soil CO\textsubscript{2}.

2 | MATERIALS AND METHODS

2.1 Carbon dioxide release experiment design

The EIT site was installed in Eumseong ~80 km away from Seoul, South Korea (36°57′44.2″ N, 127°28′03.1″ E) in 2015 and has three parts as control facility, saturated zone, and unsaturated zone (Figure 1a). Unsaturated zone of the EIT site, which is a study site targeting the CO\textsubscript{2} release test in the unsaturated zone, was divided into five zones, each of which had two release wells to inject CO\textsubscript{2} gas into the soil, with the exception that the control zone (Zone 5) had

Core ideas

- The results of this study indicated that soil CO\textsubscript{2} flux is affected by environmental factors.
- In particular, rainfall acts as a biggest inhibition factor of CO\textsubscript{2} transport.
- A permeability discrepancy leads to the vertical or horizontal flow of in-soil CO\textsubscript{2}.
- The observed correlation between CO\textsubscript{2} concentration and flux demonstrates the migration of soil CO\textsubscript{2}.
three release wells for crops, *Pinus densiflora* Siebold & Zucc. seedlings, and *Quercus variabilis* Blume seedlings (Figure 1b). Zone 1 was covered by *P. densiflora* and *Q. variabilis* seedlings, and Zones 2 and 4 were covered by sweet-potato (*Ipomoea batatas* (L.) Lam.). Thus, Zones 1, 2, 4, and 5 were designed for analyzing the effects of elevated in-soil CO₂ on physiology and growth of crops and woody-plant seedlings, whereas, Zone 3 was designed for analyzing the CO₂ migration and was grassland free of artificial interference. On the other hand, saturated zone of the EIT site is a study site targeting the CO₂ release test under the ground water level.

Perforated linear pipeline for CO₂ injection was horizontally laid at 2.5-m soil depth. From 1 to 30 June 2016, CO₂ gas was injected from the perforated release wells at Zones 1–4 with the injection rate of 6 L min⁻¹ (~15.8 kg CO₂ zone⁻¹ day⁻¹, total 1.8 t CO₂). The geophysical survey was conducted from August to September 2014. Soil bulk density was 0.68–1.08 g cm⁻³. Total N and total organic C were 0.42 g kg⁻¹ and 3.65 g kg⁻¹, respectively. pH was 5.57. The distribution of gravel (>2 mm), sand (0.063–2 mm), and silt (<0.063 mm) was 12.8, 77.6, and 9.6%, respectively. Further details on the experimental design at the EIT site were published previously (Jun et al., 2017; Kim et al., 2018).

Using the LI-8100A (LI-COR), environmental factors such as soil temperature, soil moisture, relative humidity, and atmospheric pressure at the EIT site were measured at distances of 0.0, 1.5, 3.0, 4.5, and 6.0 m from the well every 30 min in real time from 29 May to 4 Aug. 2016 (Figure 2). The soil temperature measurement was made using the thermistor, whereas soil moisture was monitored using the ECH₂O model EC-5 soil moisture probe (Decagon Devices), a 5-cm dielectric sensor for volumetric soil water content. Both soil temperature and moisture probes were installed at 5-cm depth of soil and connected directly to each flux chamber named as long-term chamber. Relative humidity inside the soil chamber and atmospheric pressure in the optical bench, which is located inside LI-8100A analyzer control unit, were measured automatically.

There were no rain events during the CO₂ injection period (1–30 June). All environmental factors showed short-term fluctuations as diurnal variations. In South Korea, summer lasts from June to August, and August is the most hot and humid month of the year. Accordingly, mean soil temperature, soil moisture, and relative humidity were 26.0 ± 6.0°C, 23.5 ± 6.8%, and 68.9 ± 15.4%, respectively, during the study period. Atmospheric pressure (98.8
2.2 Carbon dioxide concentration and flux monitoring

From 29 May to 4 Aug. 2016, measurements of CO₂ fluxes were made every 30 min. Measurements were made at the soil surface, at horizontal distances of 0, 1.5, 3.0, 4.5, and 6.0 m from the well, at the east line of Zone 3, using the LI-8100A with five long-term chambers. This automated soil CO₂ flux measurement system measured CO₂ concentration, soil temperature, soil moisture, atmospheric pressure, and relative humidity at each measurement point, as can be seen in Figure 2, and the CO₂ flux \( F_c \) was calculated as below:

\[
F_c = \frac{10VP_0 \left( 1 - \frac{W_0}{1000} \right)}{RS(T_0 + 273.15)} \times \frac{\Delta C}{\Delta t} \quad (1)
\]

where \( F_c \) is the soil efflux rate (\( \mu\)mol m\(^{-2}\) s\(^{-1}\)), \( V \) is chamber volume including the volume of the pump and measurement loop (cm\(^3\)), \( P_0 \) is the initial pressure (kPa), \( W_0 \) is the initial water vapor mole fraction (mmol mol\(^{-1}\)), \( R \) is the universal gas constant (8.314 m\(^3\) Pa K\(^{-1}\) mol\(^{-1}\)), \( S \) is the soil surface area (323.5 cm\(^2\)), \( T_0 \) is the initial air temperature (°C), and \( \Delta C/\Delta t \) is the initial rate of change in water-corrected CO₂ mole fraction (mmol mol\(^{-1}\)).

2.3 Statistical analysis

Correlation analysis was used to examine the relationships between CO₂ flux and environmental factors at 6.0-m distance from the well, and the linear relationship between CO₂ concentration and flux at five distances from the well. Also, an ANOVA was performed to compare the mean CO₂ fluxes according to distance from the well. The post-hoc test after one-way ANOVA was conducted using Duncan’s multiple range test, which is to compare every mean with every other mean. SAS 9.4 software (SAS Institute) was used for all statistical analyses.

3 RESULTS AND DISCUSSION

3.1 Soil carbon dioxide concentration and flux

Temporal variations of CO₂ concentrations and fluxes measured at the east line of Zone 3 from 29 May to 4 August...
Carbon dioxide concentrations and fluxes varied between measurement points. Carbon dioxide concentrations at 1.5 m from the well had the highest mean value at $1,279.32 \pm 416.14$ μmol mol$^{-1}$ followed by those at 0 m (933.61 $\pm$ 303.44 μmol mol$^{-1}$), 3.0 m (829.09 $\pm$ 222.20 μmol mol$^{-1}$), 4.5 m (562.62 $\pm$ 103.65 μmol mol$^{-1}$), and 6.0 m (465.78 $\pm$ 51.11 μmol mol$^{-1}$). The CO$_2$ flux measured at 0 m from the well during the injection period was $40.17 \pm 10.81$ μmol m$^{-2}$ s$^{-1}$, which was significantly lower than that measured at 1.5 m from the well as $76.40 \pm 17.30$ μmol m$^{-2}$ s$^{-1}$ ($P < .001$) (Figure 4).

Carbon dioxide concentrations and fluxes gradually increased just for ~10 d after starting the artificial CO$_2$ release but decreased sharply after CO$_2$ injection stopped. From 10 June to 1 July, CO$_2$ concentrations at all measurement points fluctuated within huge ranges, reflecting daily changes (Figure 3a). The minimum and maximum daily CO$_2$ concentration were observed during the afternoon and night times, respectively, indicating the role of the atmospheric boundary layer in soil CO$_2$ migration (Mahesh et al., 2014). However, CO$_2$ fluxes at all measurement points showed a relatively small fluctuation (Figure 3b). Carbon dioxide gas mainly migrated up to 4.5 m from the releasing point due to statistically significant differences between CO$_2$ fluxes measured at 0, 1.5, 3.0, and 4.5 m from the well and those measured at 6.0 m from the well. However, the CO$_2$ flux at 6.0 m was barely different from the background. Thus, the surface within 6.0-m radius should be monitored.

The CO$_2$ dynamics during the release experiment can be classified in two periods: a first period of steady increase of CO$_2$ fluxes, and a second period with fluctuations around a constant value (Figure 3). The CO$_2$ fluxes were measured in various distances (0.0, 2.5, 5.0, and 10.0 m) from the injection pipe with other sensors (GMP343) located at slightly different positions within Zone 3 compared with
Li-8100A measurements (Kim et al., 2018). The mean values as a function of distance from the injection well for the second period only measured using Li-8100A and GMP343 were shown in Figure 4. The peak of mean CO₂ flux was found at 2.5 m from the well for the position of GMP343 measurements, whereas this was at 1.5 m from the well for the position of Li-8100A measurements. Mean CO₂ fluxes measured using GMP343 at 0.0 and 2.5 m from the well were not significantly different as 57.32 and 58.85 μmol m⁻² s⁻¹, respectively. This trend of CO₂ migration can be directly compared with that of Li-8100A (49.05 μmol m⁻² s⁻¹ at 0.0 m and 55.51 μmol m⁻² s⁻¹ at 3.0 m from the injection well). The observed differences in the CO₂ dynamics between the two datasets illustrate the role of heterogeneity.

Diurnal variations of CO₂ concentrations and fluxes were similar, but values of concentrations and fluxes varied according to the distances from the well (Figure 5). Carbon dioxide concentrations in all measurement points had relatively high values from sunset to sunrise at all measurement points and peaked at around 0600 h. The gradual decrease and the minimum at the afternoon may result from the effects of wind and atmospheric temperature, because high wind speed has the scavenging effect on the atmospheric CO₂ concentration (Mahesh et al., 2014). Carbon dioxide fluxes in all measurement points remained about constant early in the morning and slightly increased and decreased from 0800 to 2200 h, showing only slight diurnal changes.

3.2 Effects of environmental conditions on carbon dioxide flux

The relationship between CO₂ flux and environmental factors (soil temperature, soil moisture, relative humidity, and atmospheric pressure) was analyzed to investigate the effects of the environment on CO₂ transport in soil. The correlation analysis was conducted at 6.0-m distance from the well, because it appeared that at this distance the CO₂ flux is hardly different from the background flux. Diurnal variations of environmental factors were isolated (Lewicki et al., 2010). Correlation coefficients were calculated through a 1-d moving window and presented as time series charts (Figure 6). Carbon dioxide flux showed a temporally positive correlation with soil temperature and soil moisture, and a temporally negative correlation with relative humidity and atmospheric pressure. The strongest positive and negative correlations of CO₂ flux over the entire observation period were observed with soil temperature and relative humidity (mean correlation coefficients of about .62 and -.65, respectively). The correlation coefficients of CO₂ flux with soil moisture and atmospheric pressure were .38 and -.44, respectively.
From the previous release experiment at the same site conducted in 2015, correlation between CO$_2$ flux and environmental factors was similar to the results of this release experiment conducted in 2016 (Kim et al., 2017). The previous study reported that soil temperature showed the strongest positive relationship with CO$_2$ flux ($R = .71$, $P < .0001$), so that can make the seasonal changes in CO$_2$ flux. The positive correlation between water content and CO$_2$ flux could be related to higher respiration rates with increasing water content at high temperatures (Zhang, Chen, Zhao, & Li, 2010). In this study, relatively low soil moisture appeared to be proportional to the CO$_2$ flux ($R = -.56$, $P < .0001$). On the other hand, relative humidity showed the strongest negative correlation with CO$_2$ flux ($R = -.47$, $P < .0001$). It might result from the negative relationship between temperature and relative humidity (Table 1). Atmospheric pressure also affects the diffusion of all gases including CO$_2$ from the soil to the atmosphere (Klusman, 2011; Lewicki et al., 2010). In other words, when the atmospheric pressure decreases, the CO$_2$ diffuses more rapidly from the ground into the atmosphere. Therefore, the correlation coefficient between CO$_2$ flux and atmospheric pressure was $-.15$, indicating a negative relationship ($P < .0001$). Also, correlations between CO$_2$ flux and environmental factors were analyzed during the whole experiment period (Table 1). Even though correlation coefficients were not high, CO$_2$ flux showed significant differences with all environmental factors ($P < .001$). Among the environmental factors, soil temperature and soil moisture showed the strongest positive relationship ($R = .64$, $P < .001$). The strongest negative correlation was observed between soil temperature and relative humidity ($R = -.62$, $P < .001$).

As soil gas diffusion patterns vary with environmental factors such as soil temperature, soil moisture, relative humidity, and atmospheric pressure (Kim et al., 2017), it is important to confirm which factors can accelerate or restrain CO$_2$ gas diffusion. In general, CO$_2$ flux is significantly correlated with the soil temperature-moisture interaction effect (Wildung, Garland, & Buschbom, 1975). An increase in soil temperature activates the movement of soil moisture and consequently stimulates CO$_2$ flux (Zhang et al., 2010). However, since the influence of soil temperature effects on desiccation is inversely proportional to soil moisture, CO$_2$ flux has a negative correlation with soil moisture at high soil water content (Davidson, Belk, & Boone, 1998). Atmospheric pressure also affects the rate at which CO$_2$ gas diffuses from the soil to the atmosphere.
(Klusman, 2011). It is necessary to analyze the seasonal changes of CO₂ flux through long-term monitoring, as CO₂ flux can be affected by environmental conditions, and shows seasonal changes (Görres, Kammann, & Ceulemans, 2016). Thus, even if the same amount of CO₂ leaks underground, seasonal CO₂ fluxes will be detected in different ranges on the soil surface (Kim et al., 2017).

3.3 Rainfall effects on carbon dioxide flux

There were six rainfall events, and heavy rainfall was observed from 1 to 6, 16 to 17, and 29 to 31 July (Figures 2–4). Significant changes in CO₂ fluxes at all measurement points were detected during these raining periods (Figure 3). Heavy rain had fallen in nearly all areas of the EIT site during those days, with the effect of relatively low variation of soil temperature and high variation of soil moisture and relative humidity on the EIT site (Figure 2). Filling the soil pore with water also would have suppressed the migration and diffusion of CO₂ during those days, resulting in a lower CO₂ concentration and flux (Jung, Han, Watson, Graham, & Kim, 2014). On the other hand, even though no rain was observed during the CO₂ release period, anomalously high or low CO₂ fluxes were measured from 24 to 26 June (Figure 3).

From 30 June to 6 July, the largest changes occurred in the correlation between CO₂ flux and these environmental factors due to the heavy rain (Figure 6). Also, correlation coefficients of soil temperature and moisture were slightly more or less than zero when the heavy rain events occurred on 16, 24, and 29 July. The correlation between CO₂ flux and atmospheric pressure was normally negative during the experiment period; however, this correlation coefficient sometimes showed anomalously positive values during the raining periods.

Lee, Nakane, Nakatsubo, Mo, and Koizumi (2002) reported that there was no significant correlation between CO₂ flux and soil temperature on rainy days. Heavy rain events have a significant effect on the correlation between CO₂ flux and environmental factors (Jones et al., 2014; Lewicki et al., 2010). Correlation coefficients between CO₂ flux and environmental factors showed anomalously low values or an opposite tendency (e.g., from positive to negative) due to rain events (Kim et al., 2017). In other words, the results suggested that those changes might result from the restriction of the soil permeability by the heavy rainfall, which reduced soil air-filled pore space that plays a role of gaseous CO₂ pathway (Ball, Ibert, & Jone, 1999). Thus, since heavy rain prevents the migration of CO₂ gas in the soil by acting as a physical barrier, it has a great influence on the movement of CO₂ gas (Hinkle, 1994). The unprece-
dented positive correlations between CO₂ flux and atmospheric pressure were observed during the rainfall events (Figure 6). At this time, atmospheric pressure might have decreased and CO₂ gas might dissolve in the rainfall. As a result, a positive correlation between CO₂ flux and atmospheric pressure has anomalously occurred during rainfall events.

3.4 Migration and leakage pathway of soil carbon dioxide

Characteristics of CO₂ transport at the EIT site are shown in Figure 7, which displays the correlation between CO₂ concentration and flux. During the first observation period, CO₂ gas flow measured at 0 m from the well showed a .84 correlation coefficient, which is lower than that measured at 1.5 and 3.0 m from the well (.99 and .93, respectively). Carbon dioxide gas tended to be strongly diffused at 1.5 and 3.0 m from the well, whereas low soil permeability might result in the relatively weak diffusion of soil CO₂ gas at 0 m from the well. However, during the second observation period, CO₂ fluxes at 0, 1.5, 3.0, and 4.5 m from the well showed relatively low correlation with CO₂ concentration. Thus, because of these characteristics of CO₂ gas flow in the soil, the mean CO₂ concentration and flux at 0 m from the well were lower than those at 1.5 m from the well, as shown in Figure 3. The maximum CO₂ concentrations observed were approximately 2,000, 2,300, 1,500, 1,000, and 750 μmol mol⁻¹ at 0, 1.5, 3.0, 4.5, and 6.0 m from the well, respectively; these values generally decreased as the distance from the well increased (Jones et al., 2014). The mean CO₂ concentration at 6.0 m from the well was similar to the mean CO₂ concentration attributable to natural CO₂ emission (450–500 μmol mol⁻¹); this measurement point exhibited the lowest correlation coefficient between CO₂ concentration and flux (−.21, P < .0001).

In general, soil CO₂ gas is transported within soil and from the soil to the atmosphere vertically and/or horizontally due to diffusive–advective transport (Lewicki et al., 2003). This transport of soil CO₂ gas to the surface may follow a relative increase in net soil permeability (Jung et al., 2014). Carbon dioxide transport from soil to the atmosphere indicates diffusion with vertical flow (Amundson & Davidson, 1990); however, low permeability near the soil surface may result in advection at depth with horizontal flow (Lewicki & Brantley, 2000). Those characteristics of in-soil CO₂ gas transport could be analyzed by using the correlation between CO₂ concentration and flux (Lewicki & Brantley, 2000). An area of high diffusivity with high permeability enhanced by advection at depth presents a strong positive correlation between CO₂ concentration and flux.
FIGURE 7  Correlation between CO₂ concentrations and fluxes measured at 0, 1.5, 3.0, 4.5, and 6.0 m from the well at the east line of Zone 3. Red, orange, green, blue, and purple dots in each figure indicate the data measured at the first observation period of steady increase of CO₂ fluxes, and black dots in all figures indicate the data measured at the second observation period with fluctuations around the average

An area of low diffusivity due to low permeability, enhanced by advection at depth, presents a low correlation between CO₂ concentration and flux. Naturally sourced CO₂ tends to be buoyantly transported near the surface through an area with lower soil density and viscosity than the surrounding area (Han et al., 2012). Based on the observations of CO₂ flux, the measurement point at 0 m from the well may be horizontally sealed where low-permeability soil allowed the horizontal or diagonal migration of soil CO₂ (Shipton et al., 2004). This soil property might inhibit the upward flow of deep-sourced CO₂ and may lead to a change in the leakage pathway of geologically stored CO₂ (Jung et al., 2014). Therefore, upward migration of soil CO₂ at 0 m from the well might be enhanced by greater permeability. Also, our results reflect that design of a system to detect CO₂ leakage should consider the permeability of the soil, in addition to the distance from potential sources.

4  | CONCLUSIONS

The results of the shallow CO₂ release experiment at the EIT site could provide insights into the migration of subsurface CO₂ related to a CCS leakage accident by simulating characteristics of CO₂ leakage. Carbon dioxide flux was influenced by environmental conditions. Soil moisture showed different daily pattern from relative humidity. As the results from this and previous studies indicate, relatively low soil moisture without heavy rain improved the CO₂ flux, whereas relatively high soil moisture with heavy rain would tend to block the gas pathways (even though CO₂ is soluble) and thus tend to reduce the CO₂ flux. However, relative humidity was negatively correlated with CO₂ flux. This might result from the negative relationship between temperature and relative humidity (temperature is positively correlated to CO₂ flux) rather than the direct effect of relative humidity on the CO₂ flux. The many anomalies in the temporal correlation between CO₂ flux and environmental factors observed during the raining periods indicated that rainfall may be the biggest inhibiting factor of CO₂ transport by means of closing soil pores. The interaction of low soil permeability and the buoyant nature of soil CO₂ may be one of the reasons for the intense leakage at 1.5 m from the well, compared with the other measurement points. Also, field observations of the relationship between CO₂ concentration and flux demonstrated the migration patterns of soil CO₂; low soil permeability could be an inherent characteristic of a CCS site and could result in a lateral flow of soil CO₂. Consequently, soil CO₂ would migrate not only vertically but also horizontally (or diagonally) at the subsurface, and these CO₂ movements could be detected by analyzing the relationship between CO₂ concentration and flux.

AUTHOR CONTRIBUTIONS

H.J.K. conducted field measurement and the data analysis and wrote the paper. D.K. and S.T. Y. conceived and
designed the experiments. S.H.H. and S.K. performed the experiments and analyzed the data. Y.S. supervised this research.

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CONFLICTS OF INTEREST
The authors declare no conflict of interest.

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