Development of Raman Lidar for Remote Sensing of CO₂ Leakage at an Artificial Carbon Capture and Storage Site

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Abstract: We developed a Raman lidar system that can remotely detect CO₂ leakage and its volume mixing ratio (VMR). The system consists of a laser, a telescope, an optical receiver, and detectors. Indoor CO₂ cell measurements show that the accuracy of the Raman lidar is 99.89%. Field measurements were carried out over a four-day period in November 2017 at the Eumsong Environmental Impact Evaluation Test Facility (EIT), Korea, where a CO₂ leak was located 0.2 km from the Raman lidar. The results show good agreement between CO₂ VMR measured by the Raman lidar system (CO₂ VMR_Raman LIDAR) and that measured by in situ instruments (CO₂ VMR_in-situ). The correlation coefficient (R), mean absolute error (MAE), root mean square error (RMSE), and percentage difference between CO₂ VMR_in-situ and CO₂ VMR_Raman LIDAR are 0.81, 0.27%, 0.37%, and 4.92%, respectively. The results indicate that Raman lidar is an effective tool in detecting CO₂ leakage and in measuring CO₂ VMR remotely.

Keywords: CO₂; Raman lidar; Carbon capture and storage; CO₂ leakage remote sensing

1. Introduction

The average temperature of Earth has increased by 0.8 °C since the beginning of the twentieth century, garnering worldwide interest [1]. This global warming is reported to be caused by an increase in the concentrations of greenhouse gases (e.g., CO₂, CH₄, N₂O, O₃, and CFCs) produced by human activities such as fossil fuel combustion, industrial processes, and deforestation [2,3].

As concern about increasing greenhouse gas concentrations has increased, many countries have begun to regulate carbon emissions [4] according to the Kyoto Protocol [5]. Carbon capture and storage (CCS) is considered a promising technology to reduce atmospheric CO₂ [3,6]. However, leakage of CO₂ from large-scale CCS can affect the CO₂ reduction efficiency and have serious impacts on the surrounding ecosystem, such as reduced soil pH, damage to plant and microbial communities, and groundwater contamination [7]. Previous studies [8,9] have monitored CO₂ leaking from CCS sites. Hui et al. [8] assessed CO₂ leakage from a storage facility in Xuzhou city, Jiangsu province, China, using multiple in situ instruments and wireless sensor networks. Elio et al. [9] reported a technique...
for measuring CO$_2$ soil flux at an artificial CO$_2$ injection site in Hontomin, Spain. While these in situ instruments have a high CO$_2$ measurement accuracy, many instruments are required to monitor large CCS sites. It may be difficult to detect a small, continuous CO$_2$ leak if in situ instruments are sparsely distributed at a large CCS site. In contrast, remote sensing techniques can be used to monitor CO$_2$ leakages over a large CCS site using a single instrument. However, in compiling a map of CO$_2$ distribution via two-dimensional horizontal scanning, some spots might be missed by Raman lidar because of the nature of the terrain or the presence of obstacles.

The detection limit (LDL), accuracy, and cost of the in situ instrument used by Jung et al. [10] were 0 ppm, 1.5% of reading, and about $35,000, respectively, while those of the instrument used by Jun et al. [11] were 0 ppm, 1% of reading, and about $5500, respectively. There is no commercially available Raman lidar designed to measure CO$_2$. Only one previous study [12] has reported tropospheric CO$_2$ measurement using Raman lidar, although the authors provided no information on the LDL, precision, or cost of the lidar. Atmospheric CO$_2$ measurements have been reported down to 40 ppmv for 20,000 shots at altitudes up to 1.5 km. Since a lidar system with a 50 mJ laser and repetition rate of 20 Hz costs about $40,000 [12], the cost of the lidar is thought to be higher than this figure.

In the present study, we developed a Raman lidar technique capable of detecting and remotely sensing the mixing ratio information of CO$_2$ leakage from the surface. The remainder of this paper is organized as follows. The Raman lidar hardware system and the algorithms used to retrieve the CO$_2$ volume mixing ratio (CO$_2$ VMR) are described in Section 2, and the indoor and outdoor CO$_2$ measurement performance of the system is reported in Sections 2 and 3. Finally, the discussion, and conclusions are given in Sections 4 and 5, respectively.

2. Materials and Methods

We used the elastic scattering and the Raman lidar equation to retrieve CO$_2$ mixing ratios. The lidar equation applied to CO$_2$ Raman lidar is as follows [13]:

$$P(\lambda, z) = E(\lambda) \times \exp\left( -\int_0^z a(\lambda, z)dz' \right) \times B(\lambda, z) \times \exp\left( -\int_0^z a(\lambda, z)dz' \right) \frac{S_{\text{area}}}{z^2} \times \xi(z) \times \eta_\lambda, \quad (1)$$

where $P(\lambda, z)$ is the returned backscatter signal from distance $z$ at the laser wavelength $\lambda$, $E(\lambda)$ is the initial laser energy, $a$ is the total (=aerosol + molecular) extinction coefficient, $B$ is the total (=aerosol + molecular) backscatter coefficient, $S_{\text{area}}$ is the telescope receiver area, $\xi$ describes the overlap function, and $\eta_\lambda$ is the receiver efficiency that reflects how many of the incoming photons are detected. Raman scattering refers to a process of inelastic scattering of light by molecules such as CO$_2$ in which the wavelength of the incident light is changed [14]. The Nd:YAG laser of wavelength 355 nm used in this study is shifted to a wavelength of 371.6 nm caused by the vibrational Raman process of CO$_2$ molecules. For reference, the Raman scattering wavelength of N$_2$ molecules in dry air in the lower troposphere is 386.7 nm. By using Equation (1), the backscattered signal from a Raman CO$_2$ channel at 371.6 nm, $P_{\text{CO}_2}$, can be expressed as

$$P_{\text{CO}_2}(z) = \frac{k_{\text{CO}_2}}{z^2} \sigma_{\text{CO}_2}(\pi)n_{\text{CO}_2}(z)q(\lambda_{\text{CO}_2}, 0, z) \exp\left( -\int_0^z a(\lambda, z)dz' \right), \quad (2)$$

where $k_{\text{CO}_2}$ is a proportionality constant for the system optical efficiency, the telescope receiver area, photomultiplier tube (PMT) efficiency, and the laser output energy. $\sigma_{\text{CO}_2}$ is the backscattered cross-section for CO$_2$ caused by Raman scattering, $n_{\text{CO}_2}$ is the number density for CO$_2$ as a function of distance $z$, and $a(\lambda)$ is the volume extinction coefficient at wavelength $\lambda$.

$$P_{\text{N}_2}(z) = \frac{k_{\text{N}_2}}{z^2} \sigma_{\text{N}_2}(\pi)n_{\text{N}_2}(z)q(\lambda_{\text{N}_2}, 0, z) \exp\left( -\int_0^z a(\lambda, z)dz' \right), \quad (3)$$
where \(k_{N_2}\) is a proportionality constant for the system optical efficiency, the telescope receiver area, photomultiplier tube (PMT) efficiency, and the laser output energy. \(s_{N_2}\) is the backscattered cross-section for \(N_2\) caused by Raman scattering, \(n_{N_2}\) is the number density for \(N_2\) as a function of distance \(z\), and \(a(\lambda)\) is the volume extinction coefficient at wavelength \(\lambda\).

\[
\exp\left(-\int_0^z a(\lambda, z')dz'\right) = q(\lambda_0, z_0, z),
\]

where \(q(\lambda_0, z_0, z)\) is the atmospheric transmittance from Raman lidar at distance \(z_0\) to \(z\) at wavelength 355 nm. We used Raman signals of \(N_2\) as a reference to calculate the \(CO_2\) mixing ratio. The \(CO_2\) mixing ratio is the mass of \(CO_2\) divided by the mass of dry air in a given volume [12]. Since \(N_2\) is present at a constant rate in dry air, the \(N_2\) Raman signal, \(P_{N_2}\), is used as a measure of dry air [13]. The \(CO_2\) mixing ratio can be derived from the normalization signal, which is the quotient of the \(CO_2\) Raman signal divided by the \(N_2\) Raman signal. The normalization signal can be expressed as

\[
\text{Normalization signal} = \frac{P_{CO_2}(z)}{P_{N_2}(z)}.
\]

The backscatter signals of \(CO_2\) and \(N_2\) were used to calculate the normalization signal. The Raman scattering signals of \(CO_2\) and \(N_2\) are obtained at different wavelengths (371.7 and 386.7 nm, respectively), and thus it has been assumed that there is no wavelength dependence with respect to extinction and consequently to atmospheric transmittance.

Figure 1 shows a flow chart of the process used to calculate the \(CO_2\) volume mixing ratio using the Raman lidar system. First, the normalized ratio is calculated using the \(CO_2\) and \(N_2\) inelastic signals caused by Raman scattering, as measured by the Raman lidar system.

In the right part of Figure 1, the regression coefficients are determined using the calculated normalized ratio and the \(CO_2\) VMR measured by the in situ instrument. The regression coefficients are then used in the calibration equation. The surface \(CO_2\) mixing ratios are calculated using a regression between the calculated normalized ratio and the \(CO_2\) VMR measured by an in situ instrument.

**Figure 1.** Flow chart for calculating the \(CO_2\) volume mixing ratio using the Raman lidar system.
2.1. Raman Lidar Setup

The Raman lidar system consists of the third harmonic of a Nd:YAG laser of wavelength 355 nm with 80 mJ energy and 20 Hz repetition rate, a telescope, an optical receiver, and detectors. The pulsed laser is emitted horizontally to the ground from the Nd:YAG laser. Since the laser is emitted horizontally, the diameter of the laser beam was expanded, using a beam expander, by a factor of five times to ensure eye safety. The eye safety distance of our Raman lidar system is 208 m. The Raman lidar is applied co-axially with respect to the optical system to detect CO₂ near the location of the lidar, with an overlap distance of 5–10 m. To observe CO₂ mixing ratios, the Raman lidar system measures backscatter signals caused by CO₂ Raman, N₂ Raman, and Rayleigh–Mie scattering. These signals are detected and recorded as a function of distance and wavelength. To achieve real-time monitoring, a three-channel signal collection system was designed to collect the three types of backscatter signals simultaneously (Figure 2).

![Figure 2. Schematic of the CO₂ Raman lidar system.](image)

A Schmidt–Cassegrain telescope with a diameter of 15.24 cm is used as an optical receiver. First, the scattered light is collected by the telescope. After passing through a pinhole, it is narrowed by the collimating lens (L1). A dichroic beam splitter (D.M1) reflects the light at wavelengths above 355 nm. Since the Raman scattering signal is weaker than Rayleigh–Mie scattering [12], a notch filter (F1) is used which reflects light at 355 nm and transmits it at all other wavelengths, thus reflecting the elastic signals caused by Rayleigh–Mie (total) scattering and detecting the inelastic signal caused by Raman scattering. The CO₂ filter (F2) transmits light with a wavelength of 371.7 nm to detect the Raman scattering signals of CO₂ and reflects light at all other wavelengths. The CO₂ filter (F3) transmits light at 386.7 nm and reflects all other wavelengths to detect the Raman scattering signals of N₂ needed to calculate the CO₂ VMR. The 355 nm filter (F4) transmits light at 355 nm to detect the elastic scattering signal used in aligning the lidar signal. The lenses L2, L3 and L4 are used to focus the CO₂ Raman, N₂ Raman, and elastic signals into the PMT. The signals detected by the three-channel collecting system are analyzed to determine the CO₂ mixing ratio. Table 1 summarizes the main components of the CO₂ Raman lidar system.
Table 1. Components of the Raman lidar system.

| Component | Specification |
|-----------|---------------|
| Laser     | Nd:YAG laser  |
|           | Wavelength: 355 nm |
|           | Pulse Energy: 80 mJ |
|           | Beam divergence: <1.5 mrad (full angle) |
|           | Repetition rate: 20 Hz |
|           | Pulse duration: 9 ns |
| F1        | Notch filter, 355 nm |
|           | Spectral Range: 350–1100 nm |
|           | Nominal Transmission: 1.0% |
| F2        | CO$_2$ Raman (371.7 nm) |
|           | CWL: 371.7 nm |
|           | FWHM: 0.5 nm |
|           | Tab > 70% @ 371.7 nm (Typical T > 85%) |
|           | OD > 7 @ 354.7 nm |
|           | OD > 6 from 200–1200 nm out-of-band |
| F3        | N$_2$ Raman (386.7 nm) |
|           | FWHM < 0.6 nm |
|           | OD > 7 @ 386.7 nm |
|           | Transmission: 386.7 nm > 80% T Absolute |
| F4        | 355 nm filter |
|           | CWL: 355 nm |
|           | FWHM: 10 nm |
|           | OD > 5 |
|           | Transmission > 90% |
| PMT1      | CO$_2$ Channel |
|           | R9880U-210 |
| PMT2      | N$_2$ Channel |
|           | R9880U-210 |
| PMT3      | 355 nm Channel |
|           | R9880U-210 |
| Telescope | 6-In. Schmidt Cassegrain |
| Beam expander | 5X, fixed beam-expander |
| Size of the Raman lidar system | 1100 × 850 × 550 mm (W × H × D) |
| Weight of the Raman lidar system | 70 kg |
| Power demand of the Raman lidar system | 1000 W |

2.2. Indoor CO$_2$ Cell Measurement

Indoor CO$_2$ cell measurements were carried out to quantify the accuracy of the lower detection limit of the Raman Lidar system. Figure 3 shows a schematic diagram of the indoor CO$_2$ cell setup using the Raman lidar. The cell measurement setup consists of a CO$_2$ gas vessel, a mass flow controller (MFC), a CO$_2$ gas cell, a vacuum pump, and the Raman lidar system. CO$_2$ in the CO$_2$ gas vessel is injected into the CO$_2$ cell using the MFC, which controls the CO$_2$ mixing ratio in the cell. A vacuum pump is connected to the cell to release CO$_2$ gas from the cell into the air.

To measure the VMR of CO$_2$ gas using Raman lidar, the inside of the CO$_2$ cell is evacuated and CO$_2$ gas is injected. The CO$_2$ VMR of the cell is set to range from 10% to 100%, as the lowest measurement unit of the vacuum gauge used in this study is 1%. The inelastic signals of CO$_2$ and N$_2$ caused by Raman scattering are subsequently measured by the Raman lidar and are hereafter referred to as the normalized ratio.
3. Results

3.1. Indoor CO₂ Cell Measurement

Figure 4 shows the correlation between the normalized ratio measured by CO₂ Raman lidar and the CO₂ VMRs from the CO₂ cell. The x-axis represents the CO₂ VMR inside the CO₂ cell (CO₂ VMRCELL) and the y-axis represents the normalized ratio, calculated from the inelastic signals of CO₂ and N₂ caused by Raman scattering (see Equation (5)). The correlation coefficient (R) between the CO₂ VMR_Raman LIDAR and the normalized ratio is 1, showing excellent agreement. The deviation from the regression line observed at CO₂ VMR of 10% in Figure 4 is due to the inaccuracy of the vacuum gauge used to set the CO₂ concentration in the vacuum cell. The vacuum gauge has a concentration error of 1%. Next, the CO₂ VMR in the cell is retrieved using the normalized ratio and the regression equation obtained in Figure 4 based on the method displayed in Figure 1.

Figure 4. CO₂ VMRCELL versus normalized ratio diagram.

Figure 5 shows the correlation between CO₂ VMRCELL and CO₂ VMR as measured by the Raman lidar system (CO₂ VMR_Raman LIDAR). CO₂ VMR_Raman LIDAR obtained from CO₂ Raman Lidar shows a good agreement with CO₂ VMRCELL. Both R and the slope between CO₂ VMRCELL and CO₂ VMR_Raman LIDAR are 1.00. An R value close to 1 indicates more stable and consistent laser power and higher repeatability of the detector than an R value lower than 1, because CO₂ is the only variable that changes during the training and retrieval of indoor cell measurements of CO₂. If the R value departs from 1, then the output power of the laser is not consistent or the repeatability of the detector is not
sufficient for CO\textsubscript{2} retrieval. The CO\textsubscript{2} in the cell was measured 20 times by Raman lidar under constant conditions. We calculated the error of the value measured by Raman lidar when the CO\textsubscript{2} VMR in the cell was the same as that during CO\textsubscript{2} measurements. The CO\textsubscript{2} measurement accuracy of our Raman lidar is 99.89\% based on the indoor CO\textsubscript{2} cell test.

3.2. Field Test

To examine the Raman lidar system’s capability in measuring spatially resolved CO\textsubscript{2} and N\textsubscript{2} Raman signals remotely, it was used to detect signals at a distance of 900 m from the lidar. The test was carried out on 30 October 2017, at a study site in Daejeon, South Korea, as shown in Figure 6a. We detected the Raman signals of CO\textsubscript{2} and N\textsubscript{2} for 1 h during the night. The effective spatial resolution of the Raman lidar was 250 ns (37.5 m). CO\textsubscript{2} and N\textsubscript{2} inelastic signals caused by Raman scattering were measured between the lidar system and a mountain location. During the daytime, the background signal from sunlight is stronger than the Raman scattering signal, so measurements were made only at night. Figure 6b shows the Raman signals of CO\textsubscript{2} and N\textsubscript{2} as a function of distance from the lidar system and the motorway. The increased CO\textsubscript{2} Raman signals found at 400 and 750 m from the lidar system represent the locations of a local motorway (A) and the Gapcheondosi Expressway (B), respectively. The enhanced Raman signals of CO\textsubscript{2} at locations A and B in Figure 6b imply that the lidar system is capable of detecting spatially resolved Raman signals of CO\textsubscript{2} emitted from motor vehicles. The Raman lidar obtained clear and stable Raman scattering signals of N\textsubscript{2} and CO\textsubscript{2} at a distance of 900 m from the lidar system.

Figure 5. CO\textsubscript{2} VMR\textsubscript{CELL} versus CO\textsubscript{2} VMR\textsubscript{Raman LIDAR} diagram.

Figure 6. (a) Study area in Daejeon, Korea; (b) N\textsubscript{2} and CO\textsubscript{2} Raman signals measured by the Raman lidar system.
CO₂-Leakage Measurement Campaign

To examine the CO₂-leakage measurement capability of the lidar system, measurements were carried out using both the CO₂ Raman lidar system and an in situ CO₂ probe (VAISALA, GMP343). The measurements took place between 20 and 24 November 2017, at the Eumseong Environmental Impact Evaluation Test Facility (EIT) on Seepage of Geologically Stored CO₂, an artificial CO₂ gas-leakage site (36°96′N, 127°47′E). As shown in Figure 7, CO₂ is injected at 0.5 m below the surface and left to leak back out. CO₂ gas was injected into the ground at 12 L/min for 12 h on 22 November, 2017. The Raman lidar system is located 200 m from the in situ CO₂ measurement device, which is located near the CO₂ injection location. The Raman lidar line-of-sight is located 0.5 m above the surface of the CO₂ injection inlet. The effective spatial resolution of the Raman lidar is 37.5 m and the beam diameter is about 29.98 cm at the target. The in situ instrument measured the CO₂ VMR at a distance of 1 m from the Raman lidar line-of-sight. A linear regression equation between the normalized ratio and CO₂ VMR measured by the in situ instrument is derived based on the measurements on 24 November, 2017. Six hours after stopping the CO₂ gas injection, the measurement of CO₂ on the surface was started using the CO₂ Raman lidar. The change in Raman scattering signals with respect to the change in the CO₂ mixing ratio on the surface was measured as CO₂ gas was being injected into the ground at 12 L/min. During the campaign period, the in situ instrument with its inlet located 0.5 m from the surface often failed to measure CO₂ leakage near the surface. This is thought to be associated with low CO₂ concentrations even at 0.5 m from the surface, probably due to a negligible exit velocity of CO₂ molecules. To evaluate the performance via comparison with the in situ measurement, the point of view of the lidar system was focused at the in situ inlet location to increase the lidar’s sensitivity to CO₂ molecules.

Figure 7. Schematic representation of the Eumseong Environmental Impact Evaluation Test Facility for the seepage of geologically stored CO₂.

Figure 8 shows the time series of CO₂ VMR_{in-situ} measured by the in situ instrument (blue line) and CO₂ VMR_{Raman LIDAR} measured by the Raman lidar system (red lines) at the Eumseong EIT site during the field campaign. The CO₂ gas was artificially injected into the ground and left to leak from the surface. An increasing trend in both CO₂ VMR_{in-situ} and CO₂ VMR_{Raman LIDAR} is expected with continued CO₂ injection. In Figure 8, the CO₂ VMR measured by the CO₂ Raman lidar increases steadily over time, while the CO₂ VMR measured by the in situ instrument shows an increasing trend with large variability over a short time interval. The reason for this difference between the steadily increasing CO₂ VMR_{Raman LIDAR} and the increasing CO₂ VMR_{in-situ} trend with large fluctuations could be associated with differences in measurement coverage. The spatial effective resolution of the Raman lidar is 37.5 m, which is different from the in situ instrument inlet area with a diameter of 1.5 cm. It is clearly shown that CO₂ VMR_{Raman LIDAR} tends to be lower than CO₂ VMR_{in-situ} obtained from the CO₂ leakage spot in Figure 9, which implies a certain influence of the ambient CO₂ VMR on the CO₂ VMR_{Raman LIDAR} since the ambient CO₂ VMR is likely to be lower than the CO₂ VMR of the leakage and also does not change rapidly for a short time period [15]. In the present study, if the CO₂
The system is an important factor in improving the CO₂ precision and detection limit, especially for small leaks. A high-speed photon counter, which provides fine spatial resolution, is required to enhance the precision and detection limit. The LDL and precision of the Raman lidar are also thought to affect the overall efficiency and capability of the lidar to detect such small variations in low CO₂ levels. The trend difference in Figure 8 between CO₂ VMR_{in-situ} and CO₂ VMR_{Raman LIDAR} under windy conditions could be associated with the wind direction between a CO₂ leak and the measurement location of each instrument, as the closest distance between the Raman lidar line-of-sight and the in situ instrument is 1 m. However, the effect of wind direction and speed could not be quantified due to the unavailability of wind data during the campaign period.

![Figure 8](image_url)

**Figure 8.** Time series of CO₂ VMR_{in-situ} and CO₂ VMR_{Raman LIDAR} on November 23, 2017.

![Figure 9](image_url)

**Figure 9.** CO₂ VMR_{in-situ} versus CO₂ VMR_{Raman LIDAR} diagram.

Figure 9 shows the results of linear regression for CO₂ VMR_{in-situ} and CO₂ VMR_{Raman LIDAR} at the Eumseong EIT site, revealing a correlation. The R value and slope between CO₂ VMR_{in-situ} and CO₂ VMR_{Raman LIDAR} are 0.81 and 0.15, respectively. The mean absolute error (MAE), the root mean square error (RMSE) and the percentage difference between CO₂ VMR_{in-situ} and CO₂ VMR_{Raman LIDAR}
are 0.27%, 0.37%, and 4.92%, respectively. The errors due to a decrease in backscattered Raman signals that follow a Poisson distribution are 4.9% (112.5 m), 5.4% (150.0 m), or 6.3% (187.5 m). The uncertainty due to the regression coefficients, as calculated from the residuals between the regression line and measured values, is 5.5%. Thus, the total error in CO$_2$ VMR$_{\text{Raman LIDAR}}$, which is calculated by error propagation, is estimated to be 7.4% (112.5 m), 7.7% (150.0 m), or 8.4% (187.5 m).

4. Discussion

Hui et al. [8] monitored CO$_2$ leakage from a geological CO$_2$ storage site in Xuzhou, Jiangsu, China, using multiple in situ instruments and wireless sensor networks, and Elio et al. [9] applied a technique for measuring CO$_2$ soil flux at a CO$_2$ injection site at Hontomin, Spain. While these in situ instruments have high CO$_2$ measurement accuracy, a large number of such instruments are needed to monitor large CCS sites. To enhance monitoring efficiency over a large area, remote sensing techniques are desired. Therefore, as part of efforts to develop remote sensing techniques for CO$_2$ measurements at the Earth’s surface, we developed the first compact, lightweight, and portable Raman lidar system for remote sensing of surface CO$_2$ leakage. To examine the performance of the lidar, it was used to detect strong Raman signals of CO$_2$ at two roads located 400 and 750 m from the instrument. In the case of measurements at an artificial CO$_2$ leakage site, the CO$_2$ VMRs at 0.5 m from the surface and 200 m from the Raman lidar were measured successfully and show a good correlation with data collected by an in situ instrument. It is also necessary to understand the longest remote sensing distance with the highest possible accuracy. In order to quantify the longest remote sensing distance of our Raman lidar system, it should be measured in the area without any obstacles on the line of sight. To obtain a relative accuracy from the comparison between CO$_2$ VMRs obtained from in situ instrument and the lidar, multiple in situ sensors need to be deployed at several distances on the line of the lidar sight.

During the campaign period, the CO$_2$ VMRs were detected successfully only near the surface, probably due to the low CO$_2$ exit velocity from the ground. In the case of such low velocities, the altitude of the lidar line of sight needs to be lowered to measure CO$_2$, since this compound is likely to exist near the surface. However, lowering the measurement altitude may cause noise such as fluorescence effect in the backscatter signals if the laser encounters obstacles near the surface or the ground itself, arising from the lowered altitude of the line of sight. Our current CO$_2$ retrieval algorithm needs to be improved to account for fluorescence effects. In addition, Elio et al. [9] proposed a CCS area of 3 km $\times$ 3 km that requires a Raman lidar scan that can provide sufficiently high spatial resolution to detect CO$_2$ leakage from a small site. In the present study, a laser with 80 mJ power was used in the lidar. A laser with stronger power and a more efficient receiver is needed to generate the Raman signal of CO$_2$ for sites far from the laser and to detect weak CO$_2$ Raman signals. In addition, in the case of elevated terrain along the lidar line of sight, the measurement distance on the line of sight is reduced, meaning that a lidar located at a single spot cannot complete two-dimensional horizontal scanning over a large CCS site with elevated terrain.

5. Conclusions

We developed a Raman lidar system that remotely detects CO$_2$ leakage and CO$_2$ VMR. The system consists of a laser, a telescope, an optical receiver, and detectors. In terms of indoor CO$_2$ cell measurement, CO$_2$ Raman lidar shows very high accuracy. Field measurements were carried out using Raman lidar at the Eumseong EIT site where a CO$_2$ leak is located 0.2 km from the Raman lidar system. There is good agreement between CO$_2$ VMR$_{\text{Raman LIDAR}}$ as measured by the Raman lidar system and CO$_2$ VMR$_{\text{in-situ}}$ as measured by an in situ instrument at the Eumseong EIT site. The R value and percentage difference between CO$_2$ VMR$_{\text{in-situ}}$ and CO$_2$ VMR$_{\text{Raman LIDAR}}$ are 0.81 and 4.92%, respectively. Discrepancies between the CO$_2$ values measured by Raman lidar and by the in situ instrument could be due to differences between measurement coverage and the measurement geometry of the two approaches. In the case of low CO$_2$ exit velocities, it is necessary to measure the CO$_2$ by lowering the altitude of both lidar and in situ instruments. In the case of lidar, lowering the
measurement altitude may cause noise signals such as fluorescence to be included in the backscatter signals when the laser hits the ground due to the terrain. Therefore, the lidar algorithm and measurement methods need to be studied further. The CO\textsubscript{2} VMR measurement resolution and detection limit also requires further study at low levels of CO\textsubscript{2}. In a future experiment, wind data will be used to assess the optimal measurement height for the detection and measurement of CO\textsubscript{2} leaks.

**Author Contributions:** The Raman lidar system was developed by H.K., S.C., S.P., and M.Y. In situ data collection and analysis were done by S.-T.Y. and S.-C.J. Retrieval of CO\textsubscript{2} VMR\textsubscript{Raman} was conducted by D.K., H.L., and J.-Y.R.

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