# Development of Technology for Monitoring of $CO_2$ Emissions Over Wide Areas Using a Laser System


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We are developing a system for the remote monitoring of $CO_2$ across wide areas (several kilometers) using laser technology. We have measured atmospheric $CO_2$ and that contained in combustion gases using our compact and portable test system. We were successful in measuring atmospheric $CO_2$ and that contained in exhaust gases from a combustion plant over a distance of 130 meters by using our original system that is based on a commercial laser. In this paper, we provide an outline of $CO_2$ monitoring using a laser system, and describe its innovativeness and characteristics, together with the results of $CO_2$ measurements using it.


## 1. Introduction

Today, concerns are rising that global warming may have severe impacts on the environment. The latest studies are revealing that global warming is brought about chiefly by the increase in the concentration of atmospheric $CO_2$ that is emitted in large amounts through human activities, such as combustion of fossil fuel. The Kyoto Protocol was concluded for the purpose of suppressing the increase in the concentration of atmospheric $CO_2$, or in other words, to achieve emission reduction. In all the countries of the world, mainly in advanced nations, various actions are being taken to reduce $CO_2$ in accordance with the Kyoto Protocol.

Against this backdrop, studies are being conducted on a technology (CCS) for separating and recovering $CO_2$ emitted from $CO_2$ sources, such as boilers, to store it in the earth or under the sea. This technology will fix $CO_2$ and thereby eliminate its impacts on the environment to allow $CO_2$ emissions to be regarded as being reduced. The regulation of $CO_2$ emissions has a very good chance of becoming stricter in the future so that large-scale $CO_2$ emitting facilities, such as boilers, will be obligated to recover $CO_2$.

These $CO_2$ recovery processes cannot be perfected without the means and equipment for monitoring $CO_2$ leakage over a wide area (surrounding area with a radius larger than 1 km) from the viewpoints of both safety and the environment. **Figure 1** illustrates the basic idea of our wide-area $CO_2$ leakage monitoring technology. Since $CO_2$ storage facilities are enormous in size, constant 24 hour monitoring cannot be achieved by conventional sampling methods. Even if pinpoint measurement were to be performed using detectors, a large number of monitoring devices would be required to do so, which is not practical. On the other hand, research has been conducted for many years on equipment for detecting and measuring gas components ($CO_2$, etc.) over a wide area by remote sensing technology using a laser system. Gas detection by using a laser system is generally achieved by one of the following three approaches: optical absorption, breakdown (LIBS: Laser Induced Breakdown Spectroscopy), and nonlinear optics (Raman spectrometry, multiphoton process). In $CO_2$ leakage detection, approaches based on optical absorption are used.

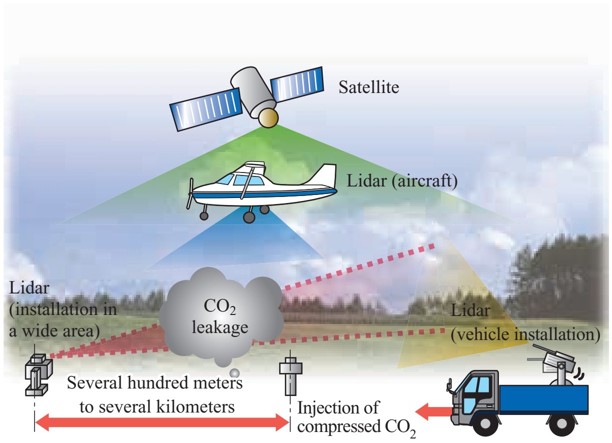



**Fig. 1** Wide-area $CO_2$ emission monitoring

**Figure 2** illustrates the principle of gas measurement by optical absorption. This method uses light that is tuned to the fine structure of the absorption wavelength of a gas such as $CO_2$. The light has a bandwidth nearly equal to the width of one of the absorption lines of the gas to

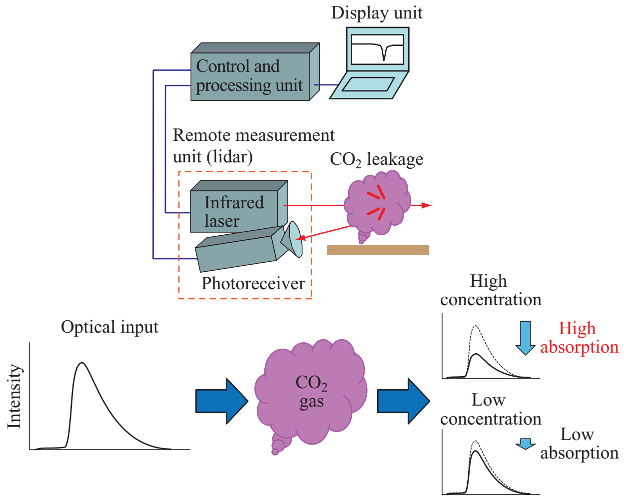



**Fig. 2** Principle of measurement of gas concentrations using optical absorption (Differential absorption)

be detected. The gas is irradiated with the light, and the scattered light is observed. In this process, the incident light is absorbed by the gas and is consequently attenuated. According to the Lambert-Beer law, transmissivity $T$ is related to gas concentration $N$, optical path length $L$ (distance that the light travels), and absorption coefficient $\alpha$ (an eigenvalue) determined by the gas species and its wavelength. The relationship is expressed by the following relational equation (1).

$$T = \exp(-\alpha NL) \quad \cdots\cdots (1)$$

This relational equation makes it possible to estimate the gas species and its concentration (integrated optical length). In addition, to eliminate the influence of other attenuating factors in the atmosphere (dust, etc.), wavelengths to be observed are generally switched between a wavelength that is absorbed (ON) by the target gas and a wavelength that is not absorbed (OFF) by the target gas, and thereby, the concentration of a specific gas can be calculated from the difference in attenuation. This method is called the differential absorption method. It requires a laser source that is tuned to a specific absorption wavelength, which differs according to gas species. However, the method is advantageous in that it allows gas concentration to be easily quantified and since it produces a higher signal intensity than other methods, it is effective in long-distance measurement.

Gas components including $CO_2$ have various absorption wavelengths in the infrared region (wavelengths larger than 0.75 μm). Technologies for measurement in the infrared region have much potential in a variety of fields, such as the environment, energy, and security. However, development of practical devices (light sources, photodetectors, etc.) for generating and detecting infrared light is behind the development of practical devices for measurement of ultraviolet and visible light. Accordingly, measurement systems based on infrared technologies have not made much progress in terms of practical use. However, studies on infrared applications have gained momentum in recent years, and infrared technologies are expected to receive more attention in fields related to development for practical use in the near future.

## 2. Issues Remaining in Wide-area $CO_2$ Measurement Using a Laser System

$CO_2$ has many absorption wavelengths in and above the near-infrared region. So, wavelengths at which absorption occurs at a practical level must be chosen while avoiding the absorption lines of water vapor, which is the greatest source of interference in wide-area (long-distance) measurement in the atmosphere. The bands that meet these conditions are the 1.6 μm band, the 2 μm band, and the 4.3 μm band. High-power laser sources and high-sensitivity photodetectors for the 1.6 μm band are readily available and are well-established as reliable hardware devices. However, the 1.6 μm band has the lowest absorption of the three wavelength bands and accordingly is disadvantageous in high range resolution and detection of trace gases. On the other hand, the 4.3 μm band has the highest absorption, so that it is advantageous in high short-range resolution and high-sensitivity detection. However, some issues remain to be solved for measurement devices in this band. For example, light sources and photodetectors have not yet been well developed, and measuring elements require cooling. From the viewpoint of the balance of the system, we used the 2 μm band, which exhibits intermediate characteristics between the two other wavelength bands.

**Figure 3** shows an example of a $CO_2$ detection system based on the differential absorption method. This system is a prototype that was experimentally constructed in order to conduct in-house verification of the measurement principle

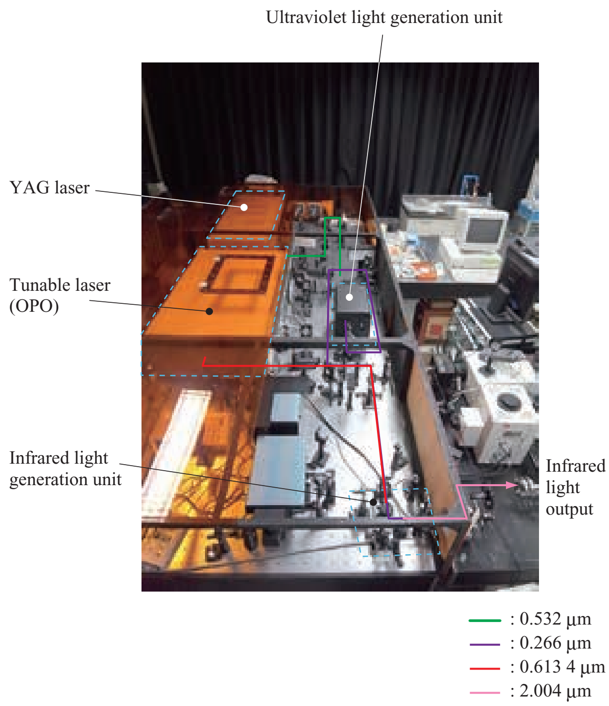



**Fig. 3** Example $CO_2$ detection system using the differential absorption method

before this study. It contains a narrow-bandwidth pulse YAG laser and an OPO (Optical Parametric Oscillator) (these two devices are commercially available). The third harmonic of the laser beam from the YAG laser excites the OPO to generate visible light (wavelength: 0.613 4 μm), and then the second harmonic of that visible light generates 0.306 7 μm ultraviolet light. In parallel, the fourth harmonic (0.266 μm) is generated by using the residual second harmonic (0.532 μm) of the laser beam generated by the YAG laser. Thereby, the system generates narrow-bandwidth light (line width: 0.04 nm (calculated value)) having a wavelength (center wavelength: approximately 2.004 μm) equal to an absorption wavelength of $CO_2$ by difference frequency generation between 0.266 μm light and 0.306 7 μm light. We used a commercially available laser unit and improved its infrared light generator to construct this system. We verified by basic laboratory testing that this system detects 100 ppm or less $CO_2$ (on a 100 m measurement distance basis). In addition, the verification revealed that it can be expected that optimization of the system, such as design improvement of its condenser lens, will achieve measurement distances of several hundred meters or longer by scaling the signal intensity of obtained data in accordance with the inverse square law of distance (according to the law, signal intensity is inversely proportional to the square of the distance).

However, such a system has the following characteristics: ① It is a large-scale, complex system and may cost several tens of millions of yen up to one hundred million yen per unit, ② Since a complex system has a high risk of failure, it is subject to the restrictions of its installation environment (temperature, humidity, etc.), and requires a skillful engineer for its maintenance, and ③ In order to cover one storage facility, anywhere from four to several tens of units may be required to be installed. Therefore, such a system is not appropriate in terms of costs acceptable for a monitoring system. Monitoring devices that have these characteristics are all designed for advanced measurement applications, so it is not effective to use these devices in the above-mentioned application. For application to leakage detection at CCS storage facilities, it is preferable to use a system that satisfies the following requirements.

- One unit of the system can cover the surrounding area with a radius larger than 1 km.
- It costs ten to twenty million yen per unit.
- It has a structure that allows site workers to easily perform maintenance work.
- It is compact and portable.
- It can detect $CO_2$ on the order of 100 ppm (on a 100 m measurement distance basis) as its minimum detectable concentration.

When the system is installed in a location and used as a fixed system, it does not need to be compact or portable; however, its installation may be subject to the restriction of the location (in terms of the size, the withstand load, etc.). In addition, the system will undergo field tests in various places. Accordingly, from the viewpoint of operation and management, it is important for the system to be compact and portable. A laser source required for realizing such a $CO_2$ measurement system is not commercially available. Therefore, it is necessary to separately develop a light source specialized for this system.

## 3. Prototype of Compact, Portable System and Result of Laboratory Test

In order to solve the issues with conventional methods, we designed[1] a wide-area $CO_2$ measuring system using a wide-bandwidth light source (on the order of several nanometers) and constructed[2] a prototype of the system. Compared with conventional systems using a narrow-bandwidth light source, the prototype is disadvantageous in detectable concentration, but it has a simple system design as a small, low-cost system. $CO_2$ is also present in the atmosphere, but when tuned for leakage detection, the system can achieve the level required for that purpose.

In order to realize this system, we developed an infrared light generator based on an OPO that operates by using a commercially available laser unit as an excitation light source. This system limits the variable wavelength range of the laser unit and operates on the premise of wide-bandwidth infrared light generation. Accordingly, it does not need to contain mechanisms for narrow-bandwidth operation. Consequently, it can be designed to have a structure specialized for $CO_2$ measurement and thereby realize a small, low-cost unit. **Figure 4** illustrates the outline of the prototype constructed for demonstrating the principle of this system. The prototype uses the second harmonic (wavelength: 0.532 μm; output: 20 mJ; pulse width: 5 ns; repetition: 10 Hz) of a commercially available pulse Nd:YAG laser as the excitation light of an OPO. This light source allows it to generate light with a wavelength of approximately 2.004 μm, which is an absorption wavelength of $CO_2$. The generated light consists of pulses that are characterized by a pulse width of 10 ns, an output up to 500 μJ, and a wavelength line width of approximately 5 nm (FWHM).

In order to evaluate the performance of the system, we conducted indoor $CO_2$ measurement (verification of detection sensitivity and verification of remote measurement). **Figure 5** outlines the results of the evaluation test. In the test, a plastic plate was placed as a diffuser 5 m away from the system, and light scattered

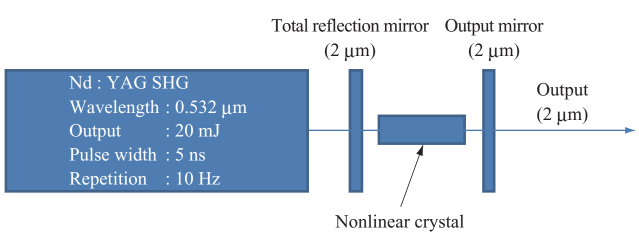



**Fig. 4** Compact and portable system for demonstration of the monitoring principle

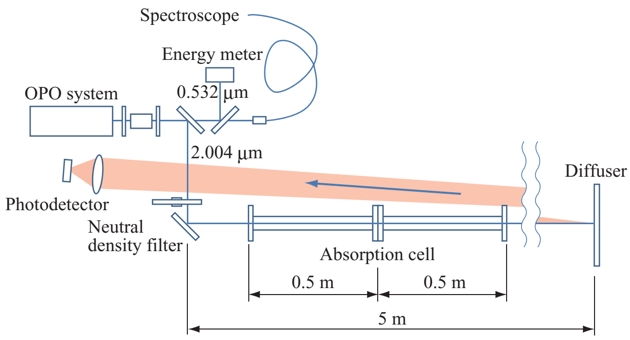



**Fig. 5** Indoor evaluation trial of system

by the plate was observed. The concentration of $CO_2$ was varied by controlling the partial pressure of $CO_2$ (balance $N_2$) in an absorption cell placed between the system and the diffuser.

**Figure 6** shows the verification result (the concentration dependency of absorption) of $CO_2$ detection sensitivity. The vertical axis denotes the logarithmic value (absorbance: $-\log T$) of absorption, and the horizontal axis denotes $CO_2$ concentration (on a 100 m measurement distance basis). As shown in **Fig. 6**, the logarithmic value of absorption is proportional to $CO_2$ concentration. This fact is consistent with the characteristics of equation (1). In addition, this test result reveals that the detection sensitivity of the system is 100 ppm or less (on a 100 m measurement distance basis). Subsequently, in order to demonstrate remote detection of $CO_2$ leakage, we conducted a remote measurement test. In the test, we observed the distance dependency of absorbance to demonstrate that the system can detect atmospheric $CO_2$. We also used pure $CO_2$ gas to demonstrate that the system can detect $CO_2$ leakage.

**Figure 7** outlines the remote measurement test and leakage detection test. We used the system to detect light scattered by a screen placed 5 to 46 m away from the system and thereby determined the distance dependency of absorbance. In addition, to simulate leakage detection, we released $CO_2$ (concentration: 100%) in an open space 5 m away from the system and thereby measured the change in absorbance. $CO_2$ continued to be released from a $CO_2$ cylinder during the measurement. **Figure 8** shows the results of the test.

When $CO_2$ was not being released, absorbance increased in proportion to distance because of the presence of atmospheric $CO_2$. Compared with the baseline of the graph of distance dependency occurring when $CO_2$ was not being released, the baseline of the graph of distance dependency occurring when $CO_2$ was being released was shifted to the high concentration side. This shift supports the fact that the system can detect $CO_2$ leakage. Absorbance was not stable when $CO_2$ was being released. This is presumably because $CO_2$ released into an open space was not stable in concentration and volume (concentration distribution).

## 4. Result of Field Test

We used this system to conduct a field test for verifying its $CO_2$ detection performance. In the test, we measured atmospheric $CO_2$ and the $CO_2$ contained in combustion gas. **Figure 9** shows an outline of the measurement test of $CO_2$

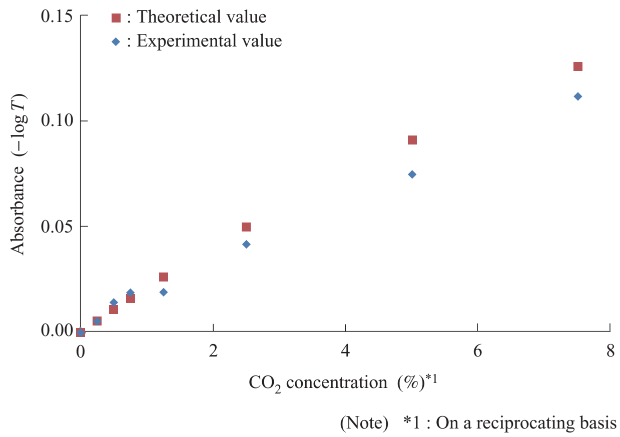



**Fig. 6** Results of verification of $CO_2$ detection sensitivity

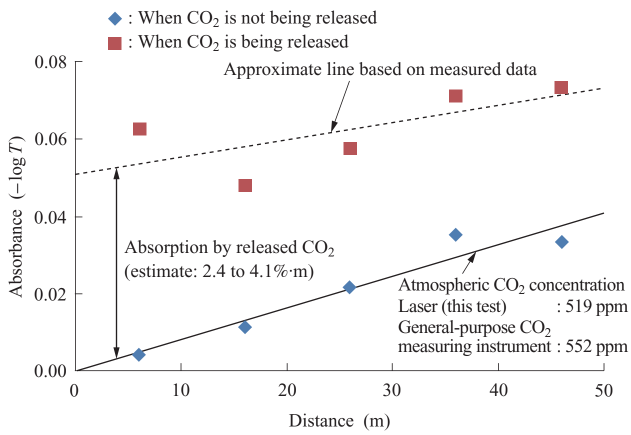



**Fig. 8** Results of remote measurement and emission detection

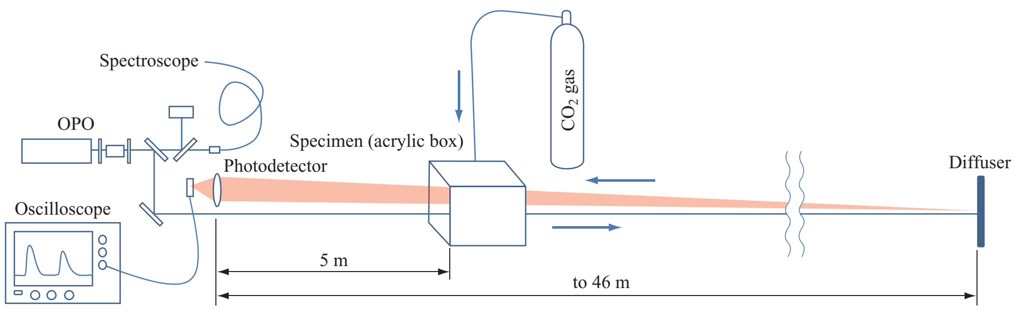



**Fig. 7** Remote measurement and emission detection

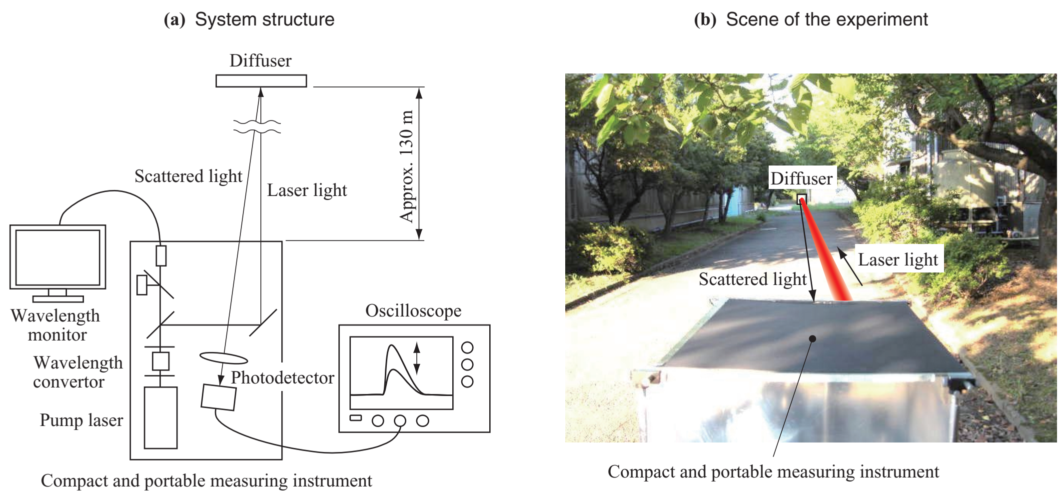



**Fig. 9** Atmospheric $CO_2$ measurement

in the outdoor atmosphere. This test was conducted in the same manner as the laboratory test of distance dependency measurement. Specifically, we radiated infrared light with a wavelength of 2 µm horizontally outdoors over a road to measure absorbance for distances up to 130 m. The properties of the infrared light were the same as those of infrared light used for the laboratory test. **Figure 10** shows

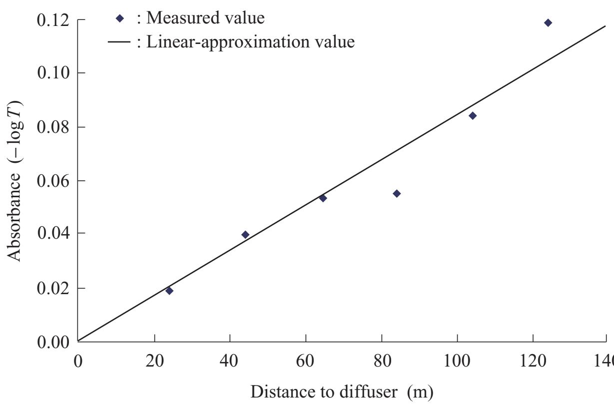



**Fig. 10** Results of atmospheric $CO_2$ measurement

the result of the test. As is the case with the laboratory test, absorbance increases linearly with the increase in distance. The absolute value of the concentration obtained from the calibration curve of the laboratory test was at the same level as the atmospheric $CO_2$ concentration. However, interfering factors (water vapor, etc.) in the outdoor environment presumably influence the results of tests. Accordingly, to verify and increase its accuracy, we will improve the system. In addition, it is presumed from signal intensity measured in the process of the test that this system has the potential to be used for measurement for a distance of 1 km or more.

We will conduct additional measurements to demonstrate that the system can be used for measurement at distances of 1 km or more. We will also determine its maximum measurement distance. In order to demonstrate the performance of outdoor $CO_2$ leakage detection by simulation and in order to examine the applicability of this system as a leakage detector at facilities such as plants, we measured the $CO_2$ contained in combustion gases. **Figure 11** shows an outline of the test.

In the test, we pointed the light generated by the

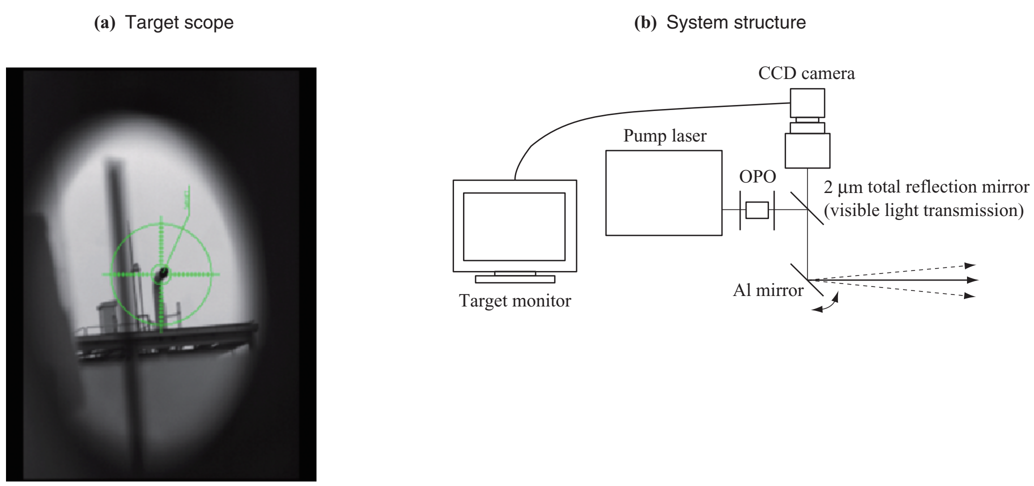
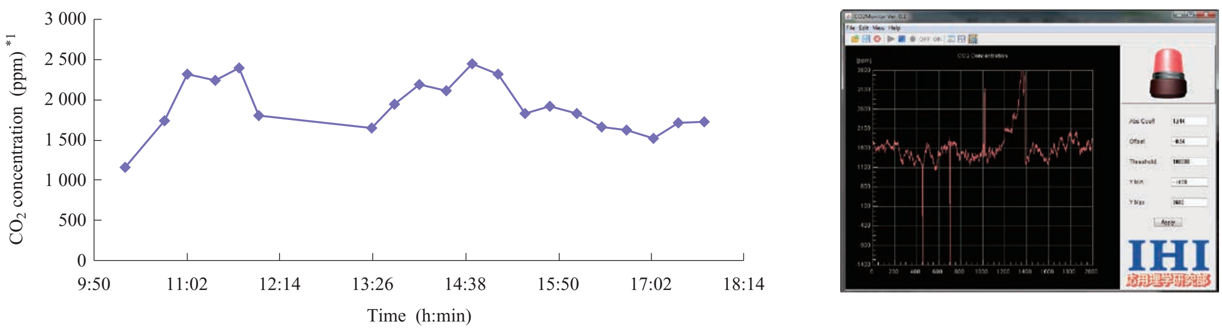



**Fig. 11** $CO_2$ measurement in combustion gases

(a) Result of manual analysis (b) Measurement screen for automatic measurement




(Note) *1 : Average value for a measurement distance of 63 m

**Fig. 12** Results of $CO_2$ measurement in combustion gases

(a) System structure (b) Appearance of the system

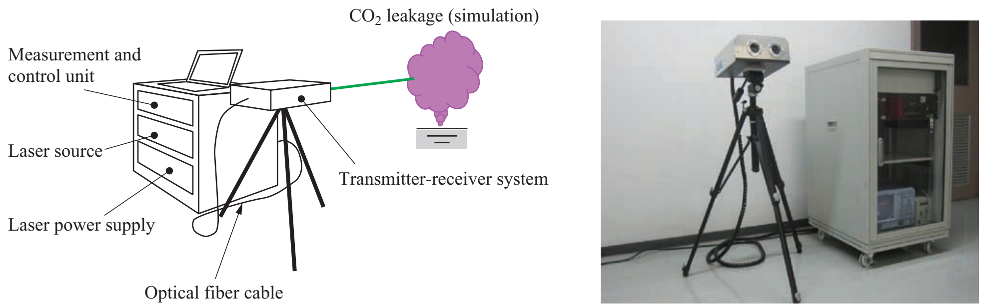



**Fig. 13** System for verification of functionality

system at an exhaust duct approximately 60 m away and observed light scattered by the duct main body. Thereby, we measured the concentration of $CO_2$ released from the duct outlet. **Figure 12** shows the result of the test. In this measurement, range resolution was not obtained, and $CO_2$ concentration was determined by integration along the optical path. Accordingly, we measured the concentration of atmospheric $CO_2$ separately and subtracted it from the measured value to determine the $CO_2$ concentration. The result was 3.15 to 12.6%·m. Since this measurement was also conducted in an open space, it is difficult to estimate the distribution of $CO_2$ concentration (the distribution will also vary depending on the effects of wind). However, it was estimated that the concentration of $CO_2$ gas released from the combustor was approximately 20%. Accordingly, the measurement result is generally consistent with the actual situation on the assumption that the combustion gas will diffuse into an area within several meters from the combustor. Therefore, we conclude that we obtained an adequate measurement result. Thus, we have demonstrated that this system can detect $CO_2$ contained in combustion gas released from a combustion plant.

## 5. Conclusions

We constructed a prototype of a wide-area $CO_2$ monitoring system using a laser unit in order to detect $CO_2$ leakage in the process of separating and recovering $CO_2$, a greenhouse gas. We also demonstrated its principle and evaluated its performance through field measurements. On the basis of the measurement results, we have concluded that the wide-area $CO_2$ monitoring system by this method has the potential for practical use.

We are now constructing a system for verifying elemental functions (**Fig. 13**) and evaluating its performance. We will improve the detection sensitivity and measurement distance of this system, demonstrate its various functions, and accumulate knowledge and information through field tests in order to put the system into practical use.

### REFERENCES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