# P2-2 Gas-Correlation Lidar System for Measuring Methane using Optical Parametric Amplifier at 3.416micro-meter

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## 1. Introduction

Atmospheric methane (1.6 ppm in the standard air) has an important roll for the global warming. Therefore development of precise measurement technique for methane is important. Measurement of this gas is also necessary for the safety. Because methane explodes when the concentration exceeds 5 %.

Most of existing laser remote-measurement techniques for methane require single mode laser or scanning of laser wavelength with high accuracy. However, precise control of laser is difficult for field measurement where vibration and change in temperature are inevitable.

We previously reported a measurement technique for atmospheric trace species using gas correlation method (Minato et al. 1998). We proposed to use a multimode laser having broad-band spectrum and a reference cell containing target gas. Single-mode lasing nor scanning of laser wavelength are not required. Therefore we can construct a measurement system which is suitable for field measurement where vibration and change in temperature are inevitable.

### 2. Development and experiment

Figure 1 shows the concept of system. Laser pulses scattered by a wall are collected by a 15-cm telescope. The round-trip optical path length is 20 m. The laser beam is divided by a beam splitter and received by two InSb detectors (EG&G J10D). A gas correlation cell containing 100% methane is put before one detector. The length of cell is 1 cm.

We developed a multimode KTA optical parametric oscillator (OPO). The OPO is pumped by a LDpumped YAG laser. It generates a signal wave of 1.5  $\mu$ m and an idler wave of 3.4  $\mu$ m. The laser power of idler wave is 10  $\mu$ J. The repetition rate is 1 kHz. The pulse width is 11 nsec. The spectral bandwidth of laser is 50 GHz which is wider than spectral band-width of absorption lines of methane.

The laser wavelength was roughly tuned monitoring signal wave by a spectrum analyzer. Then it was



Fig. 1 Gas correlation laser long-path absorption system for measuring methane.

closely tuned to the absorption lines around 3416 nm monitoring the absorption of idler wave by the methane cell.

Received laser powers by detector A and B,  $P_a$  and  $P_b,$  are expressed by

$$P_{a}(N) =$$

$$A_{a} T_{0}(t) \int P_{0}(v) \exp((N L \sigma(v))) dv, \qquad (1)$$

$$P_{b}(N) =$$

 $A_b T_0(t) \int P_0(v) \exp[(N_{cell}L_{cell}+NL) \sigma(v)] dv,$  (2) where  $A_a$  and  $A_b$  are the optical efficiencies of the measurement system including the reflectance of wall.  $T_0(t)$  represents the atmospheric turbulence effect.  $T_0(t)$ is a function of time.  $P_0(v)$  represents the spectrum of the transmitted laser. N and  $N_{cell}$  are the density of methane in the air and in the cell, respectively.  $\sigma(v)$  is the absorption cross sections of methane. L is the round-trip optical path length.  $L_{cell}$  is the length of methane cell.

Received signal decreases as optical thickness of methane increases. The laser spectrum has a component which is not absorbed by methane. Consequently, as the optical thickness of methane increases, the gradient of received signal decreases. When the  $N_{cell}L_{cell}$  is much larger than NL,  $dP_a/dN$  is much larger than  $dP_b/dN$ . From the difference between  $dP_a/dN$  and  $dP_b/dN$ , we can derive the density of methane.

Outputs of photovoltaic InSb detectors are directly connected to 1 M $\Omega$  inputs of digital oscilloscope (HP54616B). The sample rate of oscilloscope is 100 MHz. To collect data efficiently, 8 shots of signals are averaged in the digital oscilloscope. It takes about 1 second to record the averaged data.

The signal to noise ratio of developed system was evaluated first. The standard deviation of received laser power,  $\Delta P_a$ , was measured.

$$\Delta P_a/\langle P_a \rangle = 0.035$$
, (3)  
where the standard deviation is normalized by the  
average of received laser power,  $\langle P_a \rangle$ . The fluctuation  
of this value depends on the fluctuation of laser power,  
the atmospheric turbulence and the electric noise. Next,  
standard deviation of f(N), which is the ratio of  $P_a$  and

$$\Delta f(N) < f(N) > = 0.025.$$
 (4)

P<sub>b</sub>, was measured.

Here the effect of atmospheric turbulence is cancelled out. Then the methane cell is removed and the standard deviation of f(N) was measured.

$$\Delta f_{\text{NoCell}}(N) / < f_{\text{NoCell}}(N) > = 0.018.$$
(5)

Because the spectrum of each pulse changes, fluctuation of f(N) increases when methane cell is put in front of detector B.

By using a calibration cell, the error of derived density of methane was evaluated. The calibration cell, which contains 100% methane, was put in the outgoing path. The length of this cell is 1cm. Changing the pressure of methane in this cell, received signals are measured as shown in Fig. 2. In the range of density of methane as shown in Fig.2,  $P_a$  and  $P_b$  are linear with respect to optical thickness of methane. The gradient of  $P_a$  is much larger than that of  $P_b$ .

The error of derived density of methane,  $\Delta N$ , is expressed by

# $\Delta N = 1/(df(N)/dN) \Delta f(N)$ (6)

where df(N)/dN is the gradient of f(N).  $\Delta f(N)$  is the standard deviation of the ratio of P<sub>a</sub> to P<sub>b</sub>.

In Fig.2, df(N)/dN is approximately  $2.84 \times 10^{-4}$  (1/ppm · m). Because  $\Delta f(N)$  was 0.025, the error of measured density of methane is estimated to be 88 ppm · m from eq.(3). In the path length of 20 m, accuracy of 4.4 ppm is expected. This value is much lower than that of explosive level.



Fig. 2 Received signals for detector A and detector B as a function of concentration of methane in calibration cell. The density of methane in the cell is transferred to optical thickness.

To measure the methane in the background level, it is necessary to achieve higher level of accuracy. To improve the accuracy of measurement, it is necessary to extend the path length besides improving the signal to noise ratio.

## 3.Conclusion

We developed an OPO laser at 3.4  $\mu$ m and new laser long-path absorption measurement system for atmospheric methane. Because one multimode laser is used, the system is compact and stable for field measurement. As a result of experiment, it was found that the error of methane of 4.4 ppm is achieved in the round-trip path length of 20 m.

There are many absorption lines of other gas, such as  $H_2O$ ,  $CO_2$  and  $N_2O$ , in infrared range which is covered by OPO laser. The laser long-path absorption technique using the gas correlation method is easily applicable for measurements of other gases.

### References

Minato, A. Kobayashi, T. and Sugimoto, N. (1998) Laser Long-Path Absorption Lidar Technique for Measuring Methane Using Gas Correlation Method, Jpn. J. Appl. Phys. 37 3610-3613.