
NOVEL RADIO SYSTEMS
AND ELEMENTS

On a Method for Measuring Methane Concentration on Extended Atmospheric Paths Using a Remote Gas Analyzer with a Powerful Raman Amplifier

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Abstract—The paper presents the results of methane concentration measurements on extended horizontal and vertical paths in the Earth's atmosphere using a laser-generated remote gas analyzer based on a two-stage Raman amplifier with an output power of about 3 W in quasi-continuous mode with linear-frequency modulation of the laser and distributed feedback. An empirical expression of the weight function for anomalous gas distribution is obtained with a maximum concentration at a height of ~700 m and an anomalously low concentration at heights above 1000 m. This distribution agrees well with the experimental data obtained for a situational plan with additional methane emission sources. During measurements, the water absorption line was detected, which strongly depends on atmospheric humidity and can be used to refine weather data and ecological conditions.

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INTRODUCTION

In recent years, many variants of remote laser gas analyzers and methods for processing the obtained data have been developed and investigated for detecting and monitoring methane, a greenhouse gas. Three main measurement methods are distinguished: with linear frequency modulation (LFM) of the laser source with identification of the total gas absorption line [1]; with sinusoidal modulation and subsequent measurement of the second harmonic in the measured signal in the presence of a test gas on the measurement path [2]; and a pulsed method for remote measurement of methane concentration over long distances. This third method is quite common: it uses pulses with a duration of ~100 ns and a peak power of 50–90 kW, emitted by an optical parametric light generator; these pulses are received by a photodetector based on an avalanche photodiode after reflection from the target. The received light pulses at two laser frequencies in and outside the absorption line of methane make it possible to determine the integral methane concentration along the propagation path; however, the width of the methane absorption line is only determined. This method is planned to be applied in 2021 in the Merlin project for global-scale satellite methane monitoring. This project is led by two groups from the French LMD (Laboratoire de Météorologie Dynamique) and

the German Institute for Atmospheric Physics, with additional support by several research institutes in Germany and France under the aegis on the German Aerospace Center (DLR) [3]. Each of the mentioned methods has its own advantages and disadvantages in determining the spatial distribution of methane. Whereas the third, pulsed method is distinguished by large range and accuracy in measuring concentration and distance to the reflection point, the first and second methods, although possessing a sufficiently high accuracy in determining the gas concentration and the ability to record several gases at the same time, have lower accuracy in determining distance and a smaller coverage range.

The aim of this work is to create and study a gas analyzer having high accuracy in measuring gas concentration, a sufficient coverage range, and the ability to determine the distance to the reflection point, the width of the gas absorption line, and, as a result, the gas distribution in the surrounding space.

1. EQUIPMENT AND MEASUREMENT TECHNIQUE

For measurements, a prototype of a gas analyzer with an LFM of the master laser was created, followed by amplification in a high-power fiber-optic two-stage

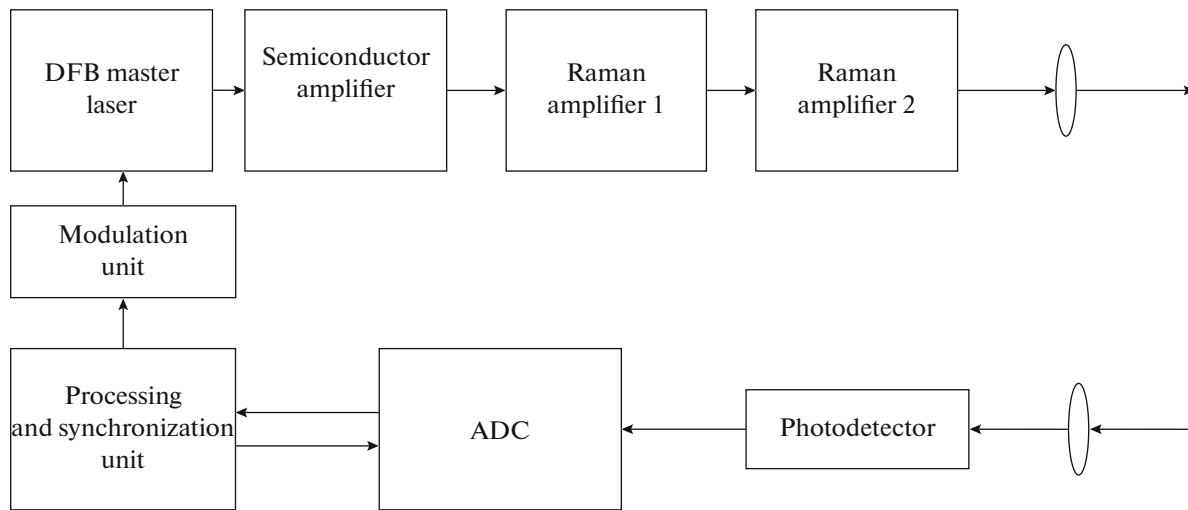


Fig. 1. Block diagram of remote gas analyzer with Raman amplifier.

Raman amplifier (Fig. 1). The transmitter at the output of the collimator emitted a power of about 3 W at a wavelength of 1653 nm. The distributed-feedback laser (OL6109L-10B DFB laser) was preamplified with a Booster Optical Amplifier (BOA-15296) semiconductor amplifier to a value of ~10 mW. A modulation unit was used for LFM of the master laser with synchronizing pulses from the processing and synchronization unit. The photodetector signal was digitized by an ADC and fed to the processing unit.

Advantages are the ability to determine both the distance to the reflection point on the trailing edge of the quasi-pulse or the correlation function [4] and the

width of the gas absorption line. The width of the absorption line can be measured quite accurately, since the entire line is obtained and digitized in the frequency scanning interval of laser radiation. This information makes it possible to specify the spatial distribution of the gas. To isolate the absorption line of the gas, calibration measurements were also carried out, when after the transmitting collimator, radiation was directed with the help of a reflecting surface through the calibration cell directly into the receiving objective, bypassing the path. Using the Bouguer law [2], it is easy to show that the formula for determining the gas concentration with this technique is

$$l = l_{\text{calibr}} - \ln\left(\frac{S}{S_{\text{calibr}}}\right)/k, \quad (1)$$

where l is the gas concentration on the path (precipitated methane layer), S is the normalized magnitude of the signal in the center of the absorption line when measured on the path, S_{calibr} is the normalized magnitude of the signal at the center of the absorption line during calibration measurements, l_{calibr} is methane deposited in the cell during calibration measurements, and k is methane absorption coefficient. If during calibration, the cell is not installed in the beam path, then $l_{\text{calibr}} = 0$ and, therefore, formula (1) is simplified.

For measurements, a horizontal path 1200 meters long and about 30 m above the Earth's surface was chosen, under which a large industrial building was being actively built, as well as a vertical path; light on this path was reflected from the clouds. In addition, two gas boilers were on the leeward side of the path, from which methane with a high background concentration may also have spread. Figure 2 shows typical images of the received signal; Fig. 3, the situational measurement plan.

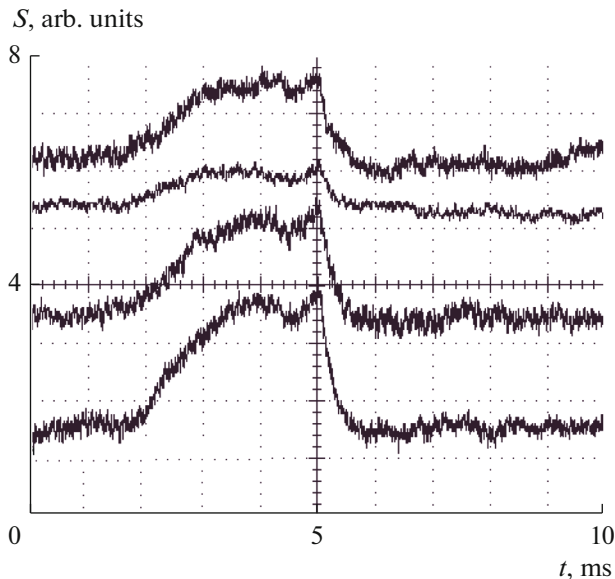


Fig. 2. Example of oscillograms of signals received from clouds with height of ~1.2–1.9 km above Earth's surface.

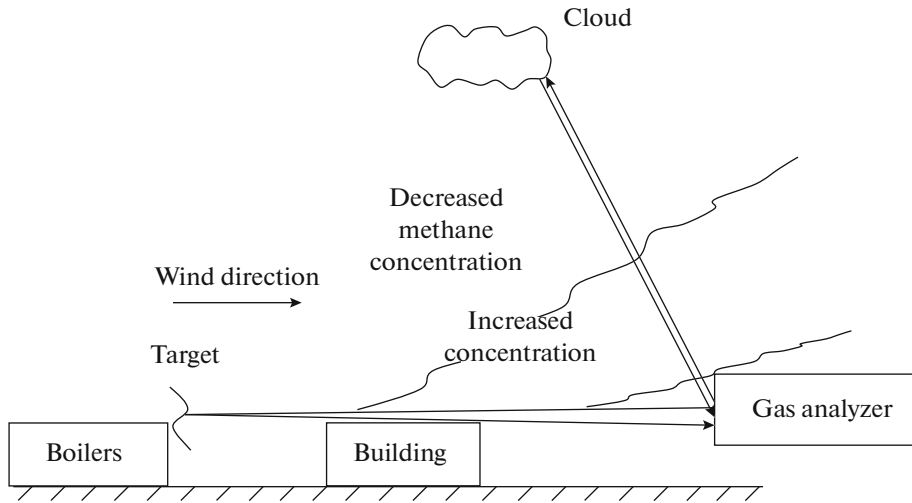


Fig. 3. Situational plan of measurements on vertical and horizontal atmospheric paths.

The signals from the photodetector were digitized in the ADC and then processed: the methane concentration, the width of the absorption line, and the distance to the reflection point were calculated. The output optical Raman amplifier worked in saturation mode, which somewhat smoothed the peak of the received LFM signal, but this had no effect on the measurement accuracy, since the calibration measurements took into account the change in shape of the emitted signal. The magnitude of the signal valley at the methane absorption line on the peak of the quasi-pulse varied with the height of the cloud layer. As a rule, the pattern is such that the higher the cloud, the smaller the received signal and the greater the relative magnitude of the valley in the gas absorption line. Five series of measurements for clouds and the horizontal path were carried out and processed. The following average values were obtained: the width of the absorption line and the deposited methane layer were equal on the horizontal path, 0.0618 nm and 2.625 mm, respectively, and on the vertical path 0.0624 nm and 3.385 mm, respectively. The theoretical values for the standard distribution of methane on the horizontal path with a standard methane background of ~2 mm/km are, respectively, 0.0618 nm and 2.40 mm at an atmospheric pressure of ~760 mmHg, and theoretical values for the standard barometric height distribution of methane yield 0.056 nm and 3.1 mm, respectively. The theoretical concentration of methane for the standard atmosphere on the vertical path was calculated by the barometric formula

$$l = \int_0^{16.5} l_0 \exp(-N/89) dN. \quad (2)$$

Integration is carried out up to $N = 16.5$ of the 100-m layer of the atmosphere (cloud height $h \sim 1.65$ km); in each 100-m layer, the methane content is $l_0 = 0.2$ mm

of the deposited methane layer. The theoretical value for the width of the absorption line at atmospheric pressure is $\gamma_0 = 0.0618$ nm [2].

2. MODEL DATA

Comparison of the experimental and theoretical data shows a difference in the results obtained. Because of this, a methane height distribution model was proposed that would satisfy the experimental data at the time of measurement. The model is based on the fact that the gas concentration was abnormally high in the atmospheric surface layer to heights of ~1000 m; at a higher height, it was below the normal value (see Fig. 3). This distribution was approximated by an empirical weight function (Fig. 4).

$$f(N) = 0.1 + 0.875 \exp\left(-\frac{N^{2.05} - 150}{160}\right) - 1.115 \exp\left(-\frac{N^{2.75} - 9.77}{100}\right), \quad (3)$$

where the current height above the Earth is determined by the number N of the 100-m layer. This function is shown in Fig. 4, whence it is clear that at the time of measurements, the methane cloud propagated upward and along the path with a concentration maximum at an height of about 700 m; above ~1000 m, the background methane concentration decreased. Based on this distribution, the model integral methane concentration was determined in accordance with weight function (3) by the formula

$$l = \int_0^{16.5} l_0 f(N) \exp(-N/89) dN. \quad (4)$$

Here, under the integral sign, l_0 corresponds, just like in (1), to the methane content in the 100-m layer of

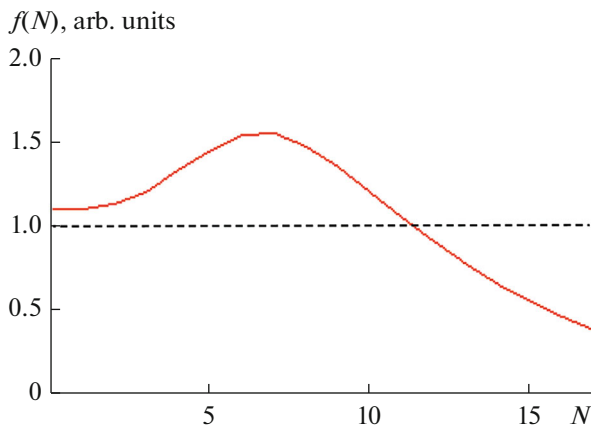


Fig. 4. Model weight function $f(Na)$ of relative height distribution of methane during measurements; N , number of 100-m of atmosphere; 17 layers correspond to height of 1.7 km; dashed line shows standard distribution.

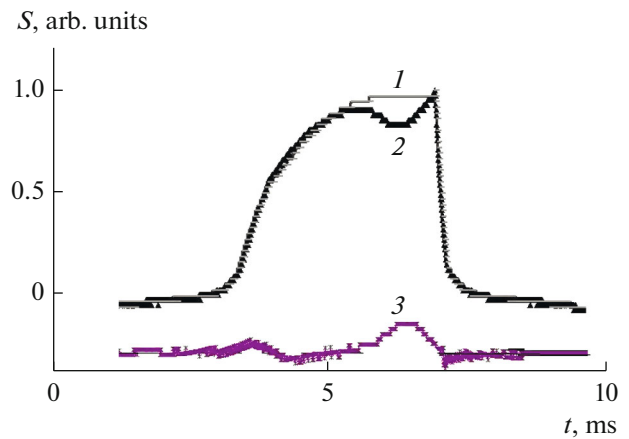


Fig. 5. Calibration waveform (1) and signal from 1.2 km path (2); difference of signals 1 and 2 (3); water absorption line is visible on left.

the atmosphere. The width of the absorption line was determined by the formula

$$\gamma = \frac{1}{16.5} \int_0^{16.5} \gamma_0 f(N) \exp(-N/89) dN, \quad (5)$$

where $\gamma_0 = 0.0618$ nm is the width of the absorption line in the surface layer. The results calculated by these model formulas, as well as theoretical and experimental results, are presented in Table 1.

As can be seen from Table 1, the line width and the deposited methane layer for the standard atmosphere differ from the measured values, and the proposed methane height distribution model (see Fig. 4) reduces this difference to the values of measurement errors and more adequately describes the real pattern of the methane cloud at the time of measurement. Differences between the modeling results and experimental data are also possible due to a small deviation in the measurement angle for clouds from 90° to the horizon.

During measurements, it was also found that in the experimental data, along with the methane absorption line, there is another gas absorption line in the left part of differential signal 3 (Fig. 5).

The differential signal is the difference between the calibration signal and the signal from the path; therefore, for accurate measurements, the calibration signal should provide a normalized signal at the photodetector output that completely replicates the signal from the path, except for areas with gas absorption lines. Otherwise the measurements will be inaccurate. As it turned out, the left peak in differential signal 3 is the water absorption line, which is separated from the methane absorption peak at ~ 0.2 nm, which is at a wavelength of 1653.7 nm. Since with increasing laser pump current, its radiation wavelength increases, the weak absorption peak corresponds to a wavelength of 1653.5 nm and is shifted by ~ 0.2 nm relative to the methane absorption line, toward smaller laser pump currents and shorter wavelengths, which completely agrees with the HITRAN database for atmospheric gas absorption in the 1653–1654 nm wavelength range (Fig. 6).

During measurements, the magnitude of the water absorption line was quite variable and strongly depended on air humidity. Simultaneous recording of the absorption lines of methane and water can be used to specify the humidity at the time of measurement, as

Table 1. Comparison of experimental, theoretical, and model values of concentrations and widths of methane absorption line on measurement paths

Source of data	Horizontal path 1.2 km		Vertical path 1.65 km	
	line width, nm	deposited layer, mm	line width, nm	deposited layer, mm
Theory	0.0618	2.4	0.056	3.1
Experiment	0.0618	2.625	0.0624	3.385
Modeling	0.0618	2.64	0.0631	3.37

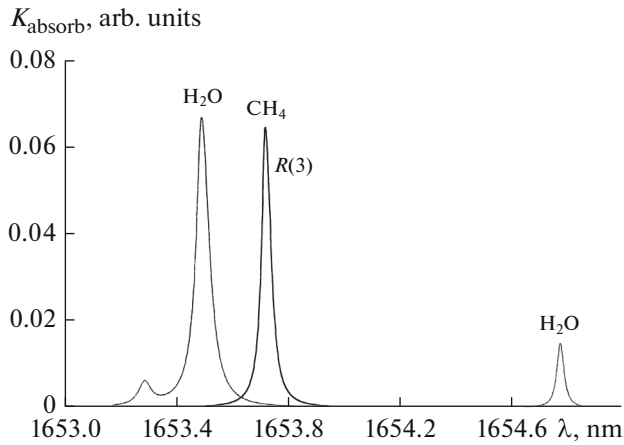


Fig. 6. Absorption spectra of CH_4 and water in atmosphere in 1653–1654 nm range according to HITRAN database.

well as for calibration measurements that refine and supplement the informational and meteorological basis of the experiment.

CONCLUSIONS

Thus, during the study, a remote laser gas analyzer based on a powerful two-stage Raman amplifier was

created and investigated, and an attempt was made to determine the spatial distribution of the background methane concentration based on measurements on two long horizontal and vertical paths. The discrepancy between the theoretical and experimental data led to refinement of the weighted height distribution function of methane based on model calculations using the standard barometric formula for the height dependence of air pressure. This technique can be useful in monitoring the methane content and distribution in the surrounding space in order to refine the background and environmental conditions.

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