Modeling and Experimental Study of Lidar Resolution to Determine Methane Concentration in the Earth's Atmosphere

V. I. Grigorievsky*a***, *, ** and Ya. A. Tezadov***^a*

*aFryazino Branch, Kotelnikov Institute of Radio Engineering and Electronics, Russian Academy of Sciences, Fryazino, Russia *e-mail: vig248@ire216.msk.su*

***e-mail: vig248@rambler.ru*

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Abstract—This article discusses the resolution of an optical lidar for measuring the background concentration of methane in the Earth's atmosphere. Using simulation and experimental studies, it has been shown that a resolution the width of the absorption line of which is ~0.006 nm and that has a concentration of methane of \sim 6% of the background can be obtained with a signal-to-noise ratio for received signals of \sim 200. When extrapolating the results to orbital measurements, the horizontal resolution will be ~8 km and the vertical resolution will be \sim 1.5 km. A broadening of the emission line of the lidar transmitter by a factor of 2–3 does not significantly reduce the resolution of the lidar, which makes it possible to increase the radiated power at the considered wavelength of ~1653 nm and improve the energy potential of the device as a whole.

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INTRODUCTION

In recent years, great interest has been shown in the global monitoring of greenhouse gases, such as methane and carbon dioxide from spacecraft, using optical methods. Particularly promising is the direction using active laser remote gas analyzers due to their high accuracy in determining the concentrations of greenhouse gas impurities. Active lidar sensing lacks the disadvantages of passive sensing methods, which require, for example, the presence of the Sun as a source and have known problems with a careful assessment of atmospheric parameters, as well as the radiative and reflective properties of the surface. The active method makes it possible to select the necessary orbits other than solar-synchronous ones. The European Space Agency (ESA) and NASA have high hopes for the use of lidars in problems of determining the content of carbon dioxide, methane, and wind speed with inaccessibility for passive methods. For instance, [1] estimates the horizontal resolution of satellite measurements of methane concentration over a fairly wide range of 50–500 km, and the error in determining the methane background will be \sim 2%, and for sounding wind speeds on a global scale from a satellite, the spatial resolution is expected to be \sim 50 km (https://www. researchgate.net/publication/234346880_The_Adm-Aeolus_Mission_the_First_Wind-Lidar_in_ Space).

The Merlin project to determine the global distribution of methane from a satellite, led by two teams from the French Laboratoire de Météorologie Dynamique and the German Institute for Atmospheric Physics, with additional support from several French and Ger-

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man research institutes at the German Center for Space and Air Flight, is planned to be implemented in 2021 (https://directory.eoportal.org/web/eoportal/ satellite-missions/m/merlin). This project considers the possibility of measuring the integral concentration of methane in an atmospheric column with an accuracy of units of percent. The measurement method is based on the differential absorption of optical radiation in and out of the methane absorption line. It is planned to use a powerful parametric light generator with a power in the pulse of $\sim 50-100$ kW in the transmitter of the gas analyzer, and a highly sensitive avalanche photodiode operating in the infrared wavelength range of \sim 1650 nm in the photodetector. The spatial resolution in the horizontal plane of such an instrument is expected to be \sim 50 km, it is not planned to determine the vertical distribution of methane in this method, since the width of the absorption line and the gas concentration, which depends on atmospheric pressure (and, therefore, height), cannot be determined by this method. To determine the vertical distribution of gas, a device is needed that would determine the width of the absorption line, as well as the distance to the reflecting point on the Earth's surface, in order to correctly solve the inverse problem [2]. That is, the active lidar equipment for determining the distribution of gas in space should probe the atmosphere from the spacecraft and, registering the signal reflected from the Earth's surface, measure the transmission of the atmosphere along the spacecraft–Earth route and back in the analytical absorption bands of controlled gases, as well as the distance to the reflection point and the width of the gas absorption line. However, the spatial and gas resolution for such an instrument remains unclear.

The aim of this work is to simulate and experimentally investigate the resolution both in space and in the concentration of a remote laser gas analyzer (lidar), which could measure not only the methane concentration, but also its absorption line width and the distance to the optical reflection point from the spacecraft signal, thereby being able to determine the distribution of methane impurity gas in space.

EXPERIMENTAL MODEL OF LIDAR AND ITS CHARACTERISTICS

In a prototype of a space lidar that could determine the vertical distribution of methane in the Earth's atmosphere, a gas analyzer can be used with linear frequency modulation (LFM) of the master laser, followed by amplification in a high-power fiber Raman amplifier [2]. Such a gas analyzer was created and tested in the laboratory and on ground tracks. A Raman amplifier based on stimulated Raman scattering in quartz fiber was used in the lidar transmitter, in which a powerful pump signal entering the active medium of the quartz fiber stimulates the Raman scattering process, which provides a shift of the pump radiation wavelength to the region of longer wavelengths and amplification of the Stokes components. In this case, the gain of the Stokes signal depends on the cross-sectional area, effective fiber length, and equivalent gain of the optical amplifier and can reach a value of about 100. The transmitter at the amplifier output emitted an optical power of about 3 W at wavelength $\lambda \sim 1653$ nm (methane absorption line *R3*) of the transmitter emission line width at a level of 3 dB was less than 0.03 nm. The distributed-feedback laser (OL6109L-10B DFB laser) was preamplified with a booster optical amplifier (BOA-15296) semiconductor amplifier to a value of \sim 10 mW. The modulation unit was the 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. In the scheme used, it is possible to determine both the distance to the reflection point from the trailing edge of the quasi-momentum [2, 3] and the width of the gas absorption line. The width of the absorption line obtained in this method can be measured quite accurately, since the entire line is received 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 gas absorption line during measurements, calibration measurements were also carried out when the radiation after the transmitting collimator of the lidar was directed directly into the receiving lens bypassing the path using a reflecting surface located next to the transceiver. In the gas analyzer, a receiving lens with a diameter of 100 mm was used, and an IAG-350 avalanche photodiode (http://www.

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lasercomponent.com) with a diameter of the sensitive area of 350 μm. To be used on a spacecraft, the parameters in terms of transmitter power and in diameter of the receiving lens of the device should be increased by approximately an order of magnitude to achieve an acceptable signal to noise ratio [4]. The forms of the signals received by the lidar, from a path length of 1200 m and calibration normalized to unity, are shown in Fig. 1. Here, the difference of these signals is shown, which can be used to judge the shape of the gas absorption line. A "dip" from the absorption line of methane present in the air is clearly visible on the signal from route 1, since the distance to the reflection point is quite large and the light passes through the gas twice as much back and forth. The signal was normalized to unity at a point *A* maximum signal, which is shifted relative to the center of the absorption line by \sim 0.0007 s in time or by \sim 0.05 nm in wavelength of the output radiation. The difference signal is a gas absorption line, which shows a sharp peak to the right of the absorption line (point *B*) that is caused by the delay of the signal from the path relative to the reference calibration signal. The time of this delay corresponds to the double measured distance to the reflection point, which is measured by the device. As can be seen from the figure, the shape of the calibration signal and the signal from the path differ from a linear one, due to the partial saturation of the transmitter amplifier, the finite width of its emission line, and also the finite integration time of the photodetector. Emission line width the transmitter was less than 0.03 nm, and the integration time of the photodetector was ~ 0.1 ms. It is undesirable to reduce these values, since the received signal will be small due to the small gain of the photodetector and the small output power of the transmitter. Both of these factors reduce the signal-to-noise ratio of the device. According to Bouguer's law, the intensity of radiation transmitted through the atmospheric layer is written in the form

$$
I/I_0 = \exp(-\alpha CL), \tag{1}
$$

where *I* is intensity of the transmitted light, I_0 is the intensity of the incident light; α is the gas absorption coefficient, *C* is methane concentration in ppm, and *L* is the length of the atmospheric path. The width of the spectral line of absorption of gas depends mainly on the pressure of atmospheric air, and absorption coefficient α (line shape) can be approximated by the Lorentz contour:

$$
\alpha(v) = N_0 \sigma(v) = N_0 \sigma(v_0) / \left(1 + \frac{(v - v_0)^2}{\gamma^2}\right), \quad (2)
$$

where N_0 is the number of molecules per unit volume $(N_0 = 2.6875 \times 10^{25} \frac{1}{m^3})$ is the Loshmidt number), σ(v) is the absorption cross section, $v = 2\pi/\lambda$ is the wavenumber (λ is the radiation wavelength), v_0 is the wavenumber at the maximum of the absorption line, and γ is the half-width of the absorption line at half

Fig. 1. Lidar waveform: (*1*) signal from track, (*2*) calibration signal, and (*3*) difference between signals 2 and 1.

Fig. 2. Normalized absorption lines for three methane background values. (*1*) 1.7, (*2*) 1.8, and (*3*) 1.9 ppm.

maximum of its amplitude. According to the HITRAN database, the width of the methane absorption line at a wavelength of 1653.73 nm, at which measurements were made, is ~0.0618 nm, and the absorption cross section at this wavelength is $\sigma(v_0) = -10^{-20}$ cm². Lidar measures L and value I/I_0 are in the range of registration bands of ~ 0.5 nm, digitizing the received signals with a clock frequency of about 1 MHz.

LIDAR RESOLUTION MODELING

Figure 2 graphically presents three normalized per unit plot dependences I/I_0 in the range of the specified wavelength range for a distance of 1200 m calculated according to formula (1) taking into account (2), and, for each of the three curves, concentration *C* that was included in (1) was varied. The methane concen-

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Fig. 3. Normalized methane absorption lines for various 2γ: (*1*) 0.06, (*2*) 0.065, and (*3*) 0.07 nm.

Fig. 4. Normalized absorption lines for three values of methane background during sensing from space orbit: (*1*) 1.7, (*2*) 1.8, and (*3*) 1.9 ppm.

trations for each of the three curves differ by 0.1 ppm for a background concentration of methane in the Earth's atmosphere equal to 1.7 ppm, this is about 6%. As can be seen from the graph, the curves at the minimum point of the methane absorption line diverge by $\sim 0.005 - 0.006$; therefore, in order for them to be distinguishable during measurements, it is necessary to ensure a signal-to-noise ratio of \sim 170–200 in the received signal. Figure 3 shows the graphs for the same sensing distance with the same concentration *C* = 1.7 ppm but different 2γ differing by 0.005 nm from each other, which is about 8% of the width of the absorption line at the half level. The graphs are also constructed according to formula (1) taking into account (2). At the same wavelength (abscissa), corre-

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sponding to approximately half the amplitude, the curves diverge by a value of $\sim 0.005 - 0.004$ arb. units on the vertical axis, therefore, to ensure the necessary sensitivity, the signal-to-noise ratio should be at least 250–200. That is, for a sensitivity (resolution) of the described lidar at 5% concentration and 8% along the width of the absorption line, the signal-to-noise ratio in the received signal should be of the order of 200– 250 units and these are rather high requirements for the "purity" of the received signal. For the signal shown in Fig. 1 and obtained over an average time of \sim 1 s, the signal-to-noise ratio is \sim 50; therefore, to reliably measure small changes in concentrations and absorption line widths for the conditions given in the experiment, an averaging time of \sim 16 s or 16 instances

of 1-s signals is required, since the signal/noise increases as a square root with increasing measurement time.

The average width of the absorption line when measured on vertical paths was determined by the barometric formula taking into account weight function $f(N)$ as

$$
2\gamma = \frac{1}{N} \sum_{1}^{N} \gamma_0 f(N) \exp(-N/89),
$$
 (3)

where $2\gamma_0 = 0.0618$ nm is the width of the absorption line in the surface layer of the atmosphere and *N* is the number of the atmospheric layer during vertical transmission. Value $f(N)$ is the weight function of the relative background distribution of methane over height. It is described by the graph in [5], from which it follows that the relative concentration of methane in the air to a height of \sim 10 km is constant and then decreases. For the surface layer of the atmosphere up to heights of $10 \text{ km } f(N) = \text{const} = 1.7 \text{ ppm}$ and formula (3) is simplified if the sensing heights do not exceed the indicated value. When calculating according to this formula, an 8% change in the absorption line width occurs when the height changes by \sim 1.5 km; therefore, we can expect that the resolution of the described lidar in height when measuring the concentration of methane will be the same value. If the integral concentrations of \sim 3 mm of the deposited layer obtained on the surface paths were measured from space orbits, then the averaging time of 16 s would allow us to obtain a horizontal resolution of \sim 128 km horizontally, if we consider the satellite speed of \sim 8 km/s and 1.5 km vertically. However, when the entire atmosphere is probed from the spacecraft, the integrated methane concentrations are much higher and the "dip" in the methane absorption line also becomes larger. Figure 4 shows the methane absorption line shapes for an effective double distance of 24 km of a horizontal path with a constant concentration of 1.7 ppm, and this distance corresponds to the integral methane content on the Earth–satellite double path. The double distance of 24 km is an order of magnitude greater than in the case of the surface track considered above. It can be seen that the "dip" on the absorption line drops to a value below 0.4; that is, its magnitude increases by more than six times compared with the case of the surface path, where the "dip" decreases from 1 to ~ 0.9 arb. units (Fig. 2). Thus, for the signals shown in Fig. 4, when the methane background concentrations are, as in the case of the surface path, 1.9, 1.8, and 1.7 ppm, the lines on the graph diverge at the minimum point by a value of \sim 0.02, rather than 0.005; i.e., that is four times larger. Because of this, for a resolution of concentration of \sim 0.1 ppm and \sim 0.004 nm along the absorption line width, the signal/noise can be four times smaller; that is, the signal accumulation time can be reduced by 16 times, that is, to 1 s. Therefore, it can be expected that the resolution of the lidar during transmission through the entire atmosphere from the spacecraft will be \sim 8 km horizontally, while the vertical resolution will remain equal to \sim 1.5 km. If averaging is performed for 16 s, then the concentration can be measured with an accuracy of \sim 1%, while the horizontal resolution with this concentration resolution will be \sim 128 km with a spatial vertical resolution of better than 0.4 km.

To increase the signal received by the lidar and increase its energy potential, it is desirable to increase the output power of the laser emitter; however, due to nonlinear effects in the fiber of the Raman converter of the optical amplifier, the radiation line width expands. The question arises of how the effect of the expansion of the emission line affects the resolution parameters of the lidar as a whole. To simulate this effect, the value of I/I_0 was calculated in the wavelength range according to the following formula:

$$
I/I_0 = \int_{\lambda-\Delta}^{\lambda} \left(\exp(-N_0 \sigma(v_0)CL / \left(1 + \frac{(v - v_0)^2}{\gamma} \right) \right) dv, \tag{4}
$$

where $\Delta = 0.08$ nm is the width of the emitted laser line; the remaining notations are the same as in formulas (1) and (2). That is, the received signals I/I_0 were actually calculated taking into account the integrating action of the transmitter radiation line, moreover, to simplify the calculations, the shape of the radiation line was assumed to be rectangular. The calculation results are shown in Fig. 5. With this integration, for a concentration of 1.7, 1.8, and 1.9 ppm, in the case of a path length of 1200 m at the wavelength of the minimum signal, the "dips" are equal to 0.935, 0.931, and 0.927, respectively. The lines on the chart diverge at the minimum point by ~ 0.004 . That is, the sensitivity to measuring the concentration decreases by ~20% compared with the case without integration. In this case, the minimum of the received signal at the absorption maximum increases somewhat. The calculation shows that a similar picture is observed in the case of methane sensing from a spacecraft, with the sensitivity to measurements also decreasing by $\sim 20\%$. A power at the output of the Raman amplifier with a line width of 0.08 nm can be obtained that is around two to three times more than in the case of a radiation line with a width of 0.03 nm; i.e., the gain in the signal-to-noise ratio can be more than twofold. Based on the results obtained, it can be said that an increase in the transmitter power, provided that the line width of its radiation does not exceed 0.08 nm, can ultimately yield a gain in the signal-to-noise ratio of more than two times and, therefore, approximately the same improvement in resolution.

RESULTS

A lidar with the characteristics presented in Section 1 operated on a horizontal path in which light was reflected from the foliage of trees and on a vertical path when light was reflected from the clouds. To test

Fig. 5. Normalized absorption lines for three values of methane background when probing on a horizontal path, taking into account the broadening of the emission line of the lidar transmitter to 0.08 nm: (*1*) 1.7, (*2*) 1.8, and (*3*) 1.9 ppm. (*4*) Emission line of transmitter. Arrow shows movement of emission line along absorption line during measurement process.

Fig. 6. Experimental results from measuring methane on tracks 600 and 300 m. (*1*) 600, (*2*) 300, and (*3*) 0 m (calibration). (*4*) Difference between signals 2 and 3. (*5*) Difference between signals 1 and 3.

the sensitivity of the lidar on a horizontal track, it was aimed at the first target at a distance of 600 m, and then at a target at a distance of 300 m. Calibration was also carried out when the target was near the transceiver. The graph in Fig. 6 presents the results. For a distance of 600 m, the concentration turned out to be 1.05 mm of the deposited layer. With a background of 1.7 ppm, the theoretical value at this distance is 1.02 mm

of the deposited methane layer. Given the measurement errors, we can say that the agreement between theoretical and experimental values is quite good. For a distance of 300 m, the deposited methane layer obtained from the measurements turned out to be 0.53 mm (the theoretical value is 0.51 mm). The difference between the obtained values of the deposited layer on the two paths amounted to 0.52 mm, which coincided with an accu-

Fig. 7. Experimental results from measuring the width of the gas absorption line on paths of 1500 and 3400 m with reflection from cloud layers: (*1*) result of averaging of 16 1-s samplings for cloud layers at a height of 3400 m; (*2*) result of averaging of 16 1-s samplings for cloud layers at a height of 1500 m.

racy of 2% with the theoretical value of 0.51 mm. As was determined earlier, to detect the difference in the background concentration of 0.1 ppm $(\sim 0.034$ mm of the deposited methane layer at a distance of \sim 340 m), it is necessary to ensure an s/n ratio of \sim 170–200. In the experiment, the signal-to-noise ratio was about 100, and this turned out to be enough to detect a difference of 0.52 mm in the deposited gas layer, which is in good agreement with theoretical modeling.

To test the simulation by measuring the width of the gas absorption line, measurements were made on a vertical path from the Earth. The reflection of the optical signal was carried out from a comparative homogeneous cloud layers located on the heights of about 3.4 km (high cloud layer) and 1.5 km (low cloud layer). Twenty-five 1-s measurements were taken of both the gas concentration on these paths and the methane absorption line width. Figure 7 shows the final measurement results. The graph shows that 16 1-s measurements are enough to measure the absorption line width with the necessary accuracy. For clarity, the graphs were smoothed using linear filtering by 16 points (extrapolating curves are shown in the same graph). It is seen that the width of the methane absorption line has a smaller width when working on a high cloud layer. The linewidth was found to be 0.054 nm, while the

theoretical value calculated by formula (4) had a value of 0.051 nm. The line width when working on a low cloud layer turned out to be 0.058 nm, while that calculated by formula (4) was 0.057 nm. As can be seen from the obtained data, the theoretical difference of 0.006 nm in the width of the absorption line was determined experimentally as 0.004 nm for 16 s. The difference between theoretical data and experimental results can be explained by some inaccuracy in determining the height of the reflection point in cloud layers and measurement errors.

CONCLUSIONS

Thus, the presented results of theoretical modeling for determining the resolution of an optical lidar both horizontally and vertically are in satisfactory agreement with the data of experimental studies. The results obtained in ground conditions show that, at a signalto-noise ratio of \sim 100–200, a difference of \sim 0.51 mm in the deposited methane layer on horizontal paths can be detected at distances of \sim 300 m, while a difference in the absorption line width of 0.006 nm can be detected at a difference in heights of ~1.9 km with an average time of \sim 16 s. Since the results of the groundbased experiment are in satisfactory agreement with the results of theoretical modeling, in measurements from the spacecraft one can also expect a satisfactory agreement with the results of modeling the resolution, which is expected to be \sim 8 km in the horizontal plane and \sim 1.5 km in the vertical plane when transmitting through the entire thickness of the atmosphere in 1 s and, respectively, 128 and 0.4 km in 16 s. Naturally, such results are possible with the corresponding energy characteristics of the lidar: approximately 30 W of the output power of the emitter and a diameter of the receiving lens of \sim 1 m. The broadening of the emission line of the lidar transmitter by two to three times makes it possible to increase the transmitter power by the same amount, which can provide a better resolution of the lidar.

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