Simulation of methane concentration measurements by lidar in a cloudy atmosphere

V I Grigorievsky and J A Tezadov
Fryazino Branch of the Institute of Radio Engineering and Electronics. V.A. Kotelnikov RAS, Russian Federation, 141190 Fryazino, Moscow region, Vvedensky sq., 1
vig248@rambler.ru

Abstract. Simulation of the vertical profile of methane concentration in the Earth's atmosphere is simulated in the absence of direct visibility on the spacecraft (SC) - Earth surface route. At high latitudes, clouds cover the Earth's surface for a significant time, so it is important to use all possibilities so that there are no significant windows of uncertainty in data acquisition along the entire path, including the lower troposphere. For modeling purposes the vertical profile of methane concentration was approximated by a sixth-degree polynomial, and the profile was refined by varying its coefficients until is coincide with the experimental data. The proposed method of using the reflected and scattered signal from clouds and cloud formations and extrapolating data along the vertical profile of methane to the entire path in the absence of direct visibility of the Earth's surface ensures uninterrupted data acquisition, thereby increasing the accuracy and reliability of statistical materials for determining the methane concentration, and, consequently, its influence on the dynamics of the climate in general.

1. Introduction
Methane is one of the active greenhouse gases in the Earth's atmosphere, influencing the formation of the planet's climate and participating in many chemical reactions in the troposphere and stratosphere [1]. After a short hiatus until 2005, the methane concentration began to increase again at a rate of ~ 6 ppb / year (parts-per-billion/year) [2]. This is due to the possible melting of permafrost, increased human economic activity, and some other factors. Monitoring methane from space can clarify forecasts on the dynamics of the development of the accumulation of methane in the atmosphere and, possibly, mitigate the consequences of global warming. Many satellites that are currently in orbits have both active and passive methane sensors on board, operating on different principles, refining the distribution of methane in space. Radio occultation measurement methods [3], original signal processing methods [4] are used, spectroscopic studies are carried out in various wavelength ranges [5, 6]. However, there are currently few active lidars as the most accurate.

The European satellite project "Merlin" to determine the global distribution of methane is planned to be implemented in 2021 [7]. This project considers the possibility of measuring the integral concentration of methane in the atmospheric column with an accuracy of a few percent. The active measurement method is based on differential absorption of optical radiation in the line and outside the absorption line of methane. It is planned to use a powerful parametric light generator with a pulse power of ~ 50-100 kW in the transmitter of the gas analyzer, and a highly sensitive avalanche photodiode operating in the infrared wavelength range of ~ 1650 nm in the photodetector.
Studies of an alternative active method with a quasi-continuous lidar on board show the possibility of using it on a spacecraft (SC). In this method the lidar emits a chirp laser beam at a wavelength of light coinciding with the gas absorption line, then receives the radiation reflected from the Earth and processes the resulting data for the purpose of determining both the integral gas concentration on the propagation path and the absorption line width.

There are often situations arise in the process of monitoring when there is no line of sight between the spacecraft and the Earth. Such situations arise due to cloudiness on the measurement path, various emissions of volcanic and other activities in the Earth's atmosphere: due to haze, fog, snowfall, etc. The question arises: is it possible under these conditions to specify the distribution of the gas concentration on the entire measurement path using the data obtained, despite the absence of direct visibility between the spacecraft and the Earth, and with what reliability?

The aim of this work is to simulate a method for reconstructing the distribution of methane concentration in the atmosphere based on lidar measurements from a space orbit and to refine the vertical profile of the background gas concentration in a cloudy atmosphere.

2. Methodology

It is known that in the case of line of sight on the measurement path, a lidar receives radiation reflected from the Earth with a power level of $\sim 10^{11} - 10^{12}$ W when using a quasi-continuous lidar with a transmitter output power of $\sim 30$ W and a receiving lens radius of $\sim 0.5$ m [8]. In this case, the signal-to-noise ratio can be $\sim 100$ when averaging the measurement results over a time of $1-10$ s when using high-sensitivity avalanche photodiodes with a detection threshold of $\sim 3.2 \times 10^{-14}$ W / Hz$^{1/2}$, which gives a horizontal resolution of $\sim 40-80$ km at a spacecraft orbital speed of $\sim 7.8$ km / s. The main sources of lidar noise are the dark current noise of the photodetector, solar illumination, and backward molecular or aerosol scattering of the atmosphere, amounting to $\sim 10^{11} - 10^{13}$ W depending on weather conditions. However, in the absence of line of sight, the received backscatter is an informative signal, which can be used to judge the vertical profile of the gas concentration in the atmosphere along the entire path up to the Earth's surface.

The quasi-continuous lidar method makes it possible to measure the averaged values of methane concentration and the width of its absorption line along the path to the point of reflection. In the case of a cloudy atmosphere this point is a cloud, aerosol, fog, etc. The proposed method for extrapolating data under the cloud layer is based on the statistical function (profile) $f(x)$ of the gas concentration distribution along the height [9]. This function allows to determine the average deposited methane layer $l(h)$ along the measurement path as an integral over the height of the product of the specified function by the barometric exponential factor of the atmospheric pressure drop with height:

$$ l(h) = \int_{0}^{h} f(75-x) \exp((75-x)/8.9) dx $$

(1)

Here $h$ is the current altitude, and the reference point $f(0)$ is taken as a distance above the Earth's surface equal to 75 km, where the function $f(x)$ vanishes. Integration is carried out up to the height $h$ towards the Earth (due to this, the distance with $h = 75$ km is the level of the Earth's surface). In the integrand is the barometric coefficient of pressure drop with height, which at $x = 75$ km (Earth's surface) becomes unity. The function $f(x)$ is well approximated by a polynomial of the sixth degree, and it looks like this:

$$ f(x) = 2.35816 \times 10^{-10}(75-x)^6 - 4.85176 \times 10^{-3}(75-x)^5 + 3.20739 \times 10^{-6}(75-x)^4 - 4.82155 \times 10^{-8}(75-x)^3 $$

$$ -0.001975395(75-x)^2 + 0.019504372(75-x) + 1.676475562 $$

(2)
Figure 1. Approximation of the statistical function $f(x)$ of methane distribution over height.

The graph of the polynomial (2) is shown on figure 1. By analogy with the barometric, this dependence is presented for the argument $(75 - x)$ so that at $x = 75$ km it turns into the average methane concentration at the Earth's surface $\sim 1.7$ ppm (molecules per million). Such an approximation of the average statistical distribution of methane over height does not differ from that presented in [9] by more than $\pm 0.01$ ppm ($\sim 0.5\%$) and it can be used with good accuracy in modeling and calculations. In turn, the height-averaged methane absorption line width is written in the following form [8]:

$$2\gamma = \frac{1}{h} \int_0^h 2\gamma_0 f'(75 - x) \exp(-(75 - x)/8.9) \, dx$$

(3)

where $f'(75-x)$ is the statistical function of the altitude distribution of methane concentration normalized to unity, and 0.0618 nm is the width of its absorption line in the surface layer of the atmosphere.

For a transparent atmosphere, based on the experimental data obtained for the average deposited methane layer and the absorption line width, it is possible to refine the standard gas distribution over the height by solving the inverse problem. If the gas concentration and the absorption line width deviate from the standard values, a distribution is sought that does not differ much from the data obtained in the experiment. In the presence of a dense cloud layer between the Earth and the spacecraft, it is possible to receive from this layer a reflected signal sufficient for registration. In this case, measurements from space orbit are considered. For example, if a cloud is located at an altitude of $\sim 10$ km above the Earth and has a refractive index $n \sim 1.3$ (water), then the reflection coefficient from it, calculated using the Fresnel formulas, is about 2%, which is comparable to the albedo of the black Earth ($\sim 2\%$), and the received reflected signal will be $\sim 3 \times 10^{-12}$ W for the above parameters of the lidar system. The cloud layer scatter signal is added to this signal. For example, if a cloud consists of droplets with a radius of $\sim 0.1$ μm (the most probable droplet size in which particles can float for a long time and not settle in the air [10]) with a density of scattering particles (density of the dispersed phase) $N_0 \sim 2 \times 10^{16}$ 1/m$^3$ at the surface Earth, then at a distance of $\sim 0.05$-0.1 m in this cloud in a solid angle of $4\pi$, there will be complete scattering of light $P_{\text{tot}}$.

This estimate was obtained on the basis of the formula for the total dissipated power in a cloud in a solid angle [11]:

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2\gamma = \frac{1}{h} \int_0^h 2\gamma_0 f'(75 - x) \exp(-(75 - x)/8.9) \, dx
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where \( P_0 = 30 \text{ W} \) is the power emitted by the lidar, \( N = 7.2 \times 10^{15} \) is the number of scattering particles at an altitude of 10 km, \( V \) is the volume of a scattering particle with a radius of \( \sim 0.1 \mu\text{m} \), \( \lambda \) is the wavelength, \( L \) is the length at which the main light scattering occurs (\( \sim 0.1 \text{ m} \)), \( n_1 \) and \( n_2 \) are the refractive indices of the dispersed phase and dispersed medium, respectively equal to 1.33 and 1. The following assumption is made in formula (4): the scattering indicatrix of a single particle is isotropic and is determined by the product of expressions in parentheses of formula (4). The interpretation of this formula consists in summing the scattered power of light from all particles located on the path of the beam in the considered layer in a solid angle of \( 4\pi \). In this case at the reception one can get \( \sim 4 \times 10^{12} \text{ W} \) of the scattered power at the specified density of scattering particles and the distance of the reflecting cloud from SC at \( \sim 440 \text{ km} \) (10 km above the Earth's surface). This level of scattered power received by the lidar \( P_{\text{scan}} \) is obtained from the following formula (5) by analogy with formula (4) for a dense cloud layer \( \sim 0.05 \text{ m} \) thick in a solid angle constricted by the receiving lens of the lidar:

\[
P_{\text{scan}} \approx P_0 \int_{R_0}^{R_0+0.05} A^2 \left( \frac{9\pi^3V^2}{\lambda^4R^2} \right) N_0 \exp\left(-\frac{(450000 - R)}{9800}\right) \frac{n_1^2 - n_0^2}{n_1^2 + 2n_0^2}^2 dR
\]

Here \( A = 0.5 \text{ m} \) is the radius of the receiving lens of the lidar, \( R_0 \) is the distance from the lidar to the scattering layer (\( \sim 440 \text{ km} \)), \( N_0 = 2 \times 10^{16} \) is the number of scattering particles near the Earth's surface, the other designations are the same as for formula (4).

The interpretation of formula (5) consists in summing the scattered power of the light falling into the receiving lens of the lidar from all particles in the path of the beam in a layer \( \sim 0.05 \text{ m} \) thick. The exponent under the integral sign reflects the barometric coefficient of concentration decrease with height.

Thus, the result of the power received by the lidar from cloud formations \( P_{\text{lid}} = P_{\text{refl}} + P_{\text{scan}} \) increases, in particular, with the number and size of scattering particles. In the considered example of the location of a dense cloud at an altitude of \( \sim 10 \text{ km} \) above the Earth it is possible to solve the inverse problem to determine the distribution function of the methane concentration with height from the cloud to the overlying layers of the atmosphere, and the condition for the continuity of the distribution function of the concentration of methane and its derivatives - to approximate and determine the distribution of the gas concentration directly under the cloud up to the surface of the Earth. The main data for such determination of the concentration are the measured values of the average width of the gas absorption line, the average deposited layer on the measured section of the path, and the distance from the spacecraft to the cloud layer. With further measurements on this segment of the spacecraft trajectory, if there is a line of sight on the path, it is possible to refine the results obtained.

3. Results and discussion

Refinement of the profile of methane concentration is reduced in the simplest case to the selection of the constant component and coefficient at the linear term of expression (2). If, for example, the measured value of the deposited layer is greater and the line width is less than the theoretical values obtained for the normal concentration profile, then the profile will correspond to curve 2 rather than curve 3, figure 2.

The figures confirming this conclusion are given below. If the cloud layer is located at a distance, for example, 10 km above the Earth, then the normal concentration profile (curve 1) corresponds to the value of the deposited methane layer 14.983 mm, and the average absorption line width is \( \sim 2.14 \times 10^3 \text{ nm} \). Let us assume that the following values were obtained in the experiment respectively: 15.93 mm and \( 2.077 \times 10^3 \text{ nm} \). It can be seen that the average deposited methane layer is about 1 mm (~ 7%) larger than for the standard profile.
Figure 2. Curve 1 - normal profile of methane concentration, 2 - concentration profile with an increased layer of deposited methane in the troposphere and a reduced layer of methane in the stratosphere (absorption line width $2.08 \times 10^{-3}$ nm), 3 - concentration profile with an increased layer of deposited methane in troposphere and stratosphere (absorption line width $2.18 \times 10^{-3}$ nm).

The selection of the coefficients at the linear and constant terms of expression (2) gives the following form of the function $f(x)$, which satisfies the experimental data:

$$f(x) = 2.35816 \times 10^{-10} (75 - x)^6 - 4.85176 \times 10^{-8} (75 - x)^5 + 3.20739 \times 10^{-6} (75 - x)^4 - 4.82155 \times 10^{-5} (75 - x)^3$$

$$-0.001975395 (75 - x)^2 + 0.015504372 (75 - x) + 1.826475562$$

(6)

The profile corresponding to curve 3 in figure 2, which differs from the standard profile only by the constant term (1.776 instead of 1.676) also gives the value of the deposited methane layer $\sim 15.98$ mm, but the absorption line width for such a curve, as shown by the calculation by formula (3), is $2.18 \times 10^{-3}$ mm and about 5% more than the measured value. For curve 2, the absorption line width is $2.08 \times 10^{-3}$ mm, which makes it a more likely distribution profile.

For the case of clouds such as fog, when there are “loose” formations in the air, for example, from small water droplets, the received signal can also be calculated based on Rayleigh backscattering for particles small compared to the wavelength or Mie scattering (at the lidar radiation wavelength comparable of scattering particles). For example, if the distance at which total light scattering occurs is $\sim 10$ km above the Earth's surface, then the density of particles of a “loose” cloud (or fog) should be $\sim 10^5$ times less than in the above case of a dense cloud $N_0 \sim 2 \times 10^{16}$ 1/m$^3$, that is, $N_0 \sim 2 \times 10^{11}$ 1/m$^3$ of particles with a radius of $\sim 0.1$ μm. In this case, scattering occurs in the entire 10 - km layer of fog up to the Earth's surface, mainly in the lower layer $\sim 5$-10 km long. Since the result of the received scattered power strongly depends on the density and size of the scattering particles, it is desirable to have information on the composition of the scattering medium during sensing, which can be used, for example, a synchronous photograph of the measurement path and the magnitude of the received signal. In the case of “loose” fog (the concentration of particles at the Earth's surface is $N_0 \sim 2 \times 10^{11}$ 1/m$^3$), the power $P_{lid}$, received by the lidar, from the layer of the atmosphere 5000 m thick, located at a distance $R_0$ from the lidar, can be determined by a formula similar to (5):
\[ P_{\text{lid}} = \int_{R_0}^{R_0+5000} P_0 \pi A^2 \left( \frac{9\pi^2 V^2}{\lambda^4 R^2} \right) N(\exp((450000 - R) / 9800))(\frac{n_1^2 - n_0^2}{n_1^4 + 2n_0^2})^2 dR, \]  

where the designations are the same as for formula (5). Here, the scattered power of light entering the receiving lens of the lidar from particles located in the path of the beam in a layer \( \sim \) 5000 m thick is summed up. The power received by the lidar and calculated by formula (7) is graphically shown in figure 3.

\[ P_{\text{lid}} \times 10^{12}, \text{ W} \]

![Graph](image)

Figure 3. The power entering the receiving lens of the lidar for a scattering layer of 5000 meters.

It can be seen from the figure 3 that the main source of the lidar signal is the lower atmosphere with a height of \( \sim \) 10 km. Let's define the most probable area of the laser beam reflection as the center of gravity of the figure bounded by the graph. It is easy to calculate using (7) that this point is located at a distance of \( \sim \) 7 km from the Earth's surface. Thus, assuming that the lidar has a distance sensor that, in the case of loose fog, gives an indefinite distance to the reflection region due to a significant spread in the arrival time of the reflected pulse, we can consider a reliable reflection region as a distance of \( \sim \) 7 km above the Earth's surface. It is desirable to estimate the distance to the point of reflection with an accuracy of \( \sim \) 1000 meters. In this case, the results of approximating the concentration profile are obtained more accurately. In quasi-continuous lidars with direct photodetection of received signals, it is possible to determine the distance to the reflected object, for example, by the trailing front of the received quasimomentum, but in the case of “loose” fog, such a measurement gives large errors, on the order of several kilometers. The distance to the reflection point is entered equal to 7 km above the Earth's level (443 km from the spacecraft orbit).

When considering the option of cloudiness intermediate between the above-considered extreme cases, when dense clouds are in the lower layer of the troposphere or there is not “loose” fog, but fog with a higher concentration of droplets, the distance to the reflection region can be measured by a distance sensor within the specified accuracy \( \sim \) 1000 meters and this is enough to refine the profile of methane concentration along the entire route also with a resolution of \( \sim \) 1000 meters.

4. Conclusions
The modeling of the distribution of the vertical profile of the concentration of methane in the Earth's atmosphere in the absence of direct visibility on the spacecraft flight trajectory - the Earth's surface is carried out. The proposed method of using the reflected and scattered signal from clouds and aerosol formations and extrapolating data along the vertical profile of methane to the entire path in the absence of direct visibility of the earth's surface allows eliminating the lack of information and increasing the
accuracy and reliability of statistical data for determining the concentration of methane, and, hence, also of its influence on the dynamics of the climate as a whole.

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